Supporting Information
CarbX - A programme for the evaluation of wide-angle X-ray scattering data of non-graphitic carbons

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1 Mathematical Background

1.1 Calculation of the Atomic Scattering Factors

The atomic scattering factors for carbon $f_{\mathrm{C,perp}}$ (perpendicular to the graphene sheets) and $f_{\mathrm{C,para}}$ (parallel to the graphene sheets) are calculated by the following interpolation functions:

$$\begin{aligned} f_{\mathrm{C,perp}} = 0.289677 &+ 1.05075 \cdot \exp\left(-2.43905 \cdot s^2\right) \\ &+ 1.56165 \cdot \exp\left(-0.164166 \cdot s^2\right) \\ &+ 3.09995 \cdot \exp\left(-28 \cdot s^2/4\right) \end{aligned} \tag{1}$$

where $s = (2 \cdot \sin(\theta)/\lambda)$ is the modulus of the scattering vector in 1/Å, θ the scattering angle in degrees and λ the wavelength used for the measurement in Å.

$$\begin{aligned} f_{\mathrm{C,para}} = 0.289677 &+ 1.05075 \cdot \exp\left(-2.43905 \cdot s^2\right) \\ &+ 1.56165 \cdot \exp\left(-0.164166 \cdot s^2\right) \\ &+ 3.09995 \cdot \exp\left((-28 - \Delta_{\mathrm{an}}) \cdot s^2/4\right) \end{aligned} \tag{2}$$

The atomic scattering factors for nitrogen f_{N} and oxygen f_{O} are obtained by interpolating of the values given *International Tables for X-ray Crystallography* as depicted in Figs S1 and S2.

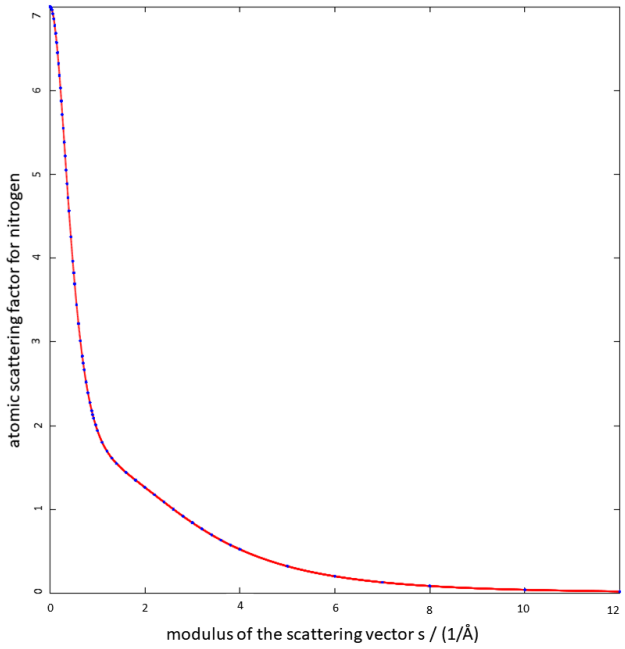

Figure S1: Interpolation of the the atomic scattering factor of nitrogen (red) along with the data points used for the interpolation (blue).

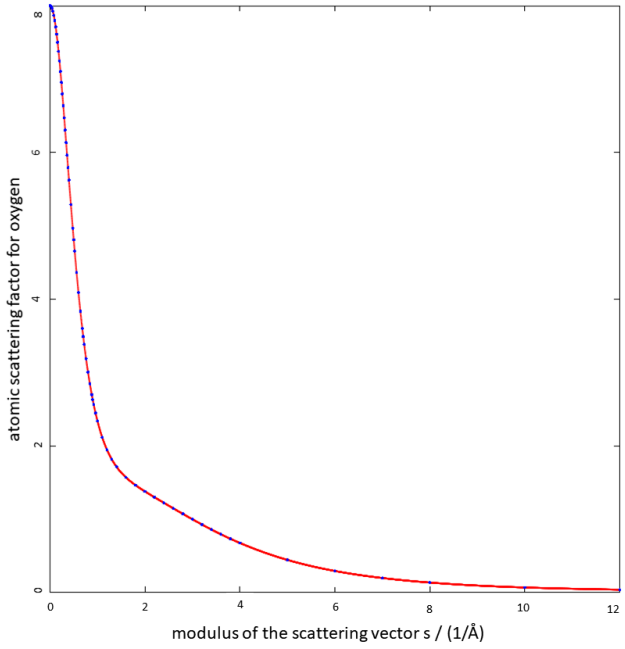

Figure S2: Interpolation of the the atomic scattering factor of oxygen (red) along with the data points used for the interpolation (blue).

1.2 Interlayer scattering

The interlayer scattering I_{inter} is given by

$$\begin{aligned} &I_{\text{inter}}\left(L_{\text{c}}, \kappa_{\text{c}}, a_3, a_{3,\min}, \sigma_3, u_3, \eta, l_{\text{cc}}, q, s\right) \\ &= \frac{n_0 \cdot g_0\left(q\right)}{2 \cdot \pi \cdot s^2 \cdot S_0\left(l_{\text{cc}}\right)} \cdot \left[1 + \eta \cdot D\left(u_3, s\right) \cdot \left[I_{\text{1D}}\left(L_{\text{c}}, \kappa_{\text{c}}, a_3, a_{3,\min}, \sigma_3, s\right) - 1\right]\right] \end{aligned} \tag{3}$$

where $n_0 = 2$ is number of atoms per unit cell of the 2D graphene layer lattice, g_0 describing the preferred orientation of the stacks, $2 \cdot \pi \cdot s^2$ for spherical averaging, $S_0\left(l_{\text{cc}}\right) = 3^{3/2} \cdot l_{\text{cc}}^2/2$ the surface of the unit cell, D describing the thermal motion and I_{1D} the normalized interference function of a disordered 1D lattice (Faber, 2014).

The preferred orientation of the stacks is described by g_0 as in (Ruland and Smarsly, 2002):

$$g_0\left(q\right) = \frac{\left(1+q\right) \cdot \sqrt{q}}{\operatorname{arctanh}\left(\sqrt{q}\right) \cdot \left(1-q\right)^2} \tag{4}$$

D, describing the thermal motion is given, by

$$D\left(u_3, s\right) = \exp\left(-4 \cdot \pi^2 \cdot u_3^2 \cdot s^2\right) \tag{5}$$

(Ruland and Smarsly, 2002) & (Faber, 2014).

The normalized interference function of a disordered 1D I_{1D} lattice is is calculated as elucidated in (Ruland and Smarsly, 2002) & (Faber 2014):

$$\begin{aligned} &I_{\text{1D}}\left(L_{\text{c}}, \kappa_{\text{c}}, a_3, a_{3,\min}, \sigma_3, s\right) = \\ &\operatorname{Re}\left[\frac{1 + H\left(a_{3,\min}, a_3, \sigma_3, s\right)}{1 - H\left(a_{3,\min}, a_3, \sigma_3, s\right)} - \frac{2 \cdot H\left(a_{3,\min}, a_3, \sigma_3, s\right) \cdot \left(1 - \left\langle H^N\right\rangle\left(L_{\text{c}}, \kappa_{\text{c}}, a_{3,\min}, a_3, \sigma_3, s\right)\right)}{\frac{L_{\text{c}}}{a_3 \cdot (1+\kappa_{\text{c}})} \cdot \left(1 - H\left(a_{3,\min}, a_3, \sigma_3, s\right)\right)^2}\right] \\ &- \frac{1}{a_3} \cdot I_0\left(L_{\text{c}}, \kappa_{\text{c}}, a_{3,\min}, a_3, s\right) \end{aligned} \tag{6}$$

where H is defined as

$$H\left(a_{3,\min}, a_3, \sigma_3, s\right) = \frac{\exp\left(2 \cdot \pi \cdot i \cdot a_{3,\min} \cdot s\right)}{\left(\frac{1 - 2 \cdot \pi \cdot i \cdot (\sigma_3)^2 \cdot s}{a_3 - a_{3,\min}}\right)^{\left(\frac{a_3 - a_{3,\min}}{\sigma_3}\right)^2}}, \tag{7}$$

$\left\langle H^N\right\rangle$ is defined as

$$\begin{aligned} &\left\langle H^N\right\rangle\left(L_{\text{c}}, \kappa_{\text{c}}, a_{3,\min}, a_3, \sigma_3, s\right) \\ &= \left(1 - \frac{L_{\text{c}} \cdot \kappa_{\text{c}}}{a_3 \cdot (1+\kappa_{\text{c}})} \cdot \log\left(H\left(a_{3,\min}, a_3, \sigma_3, s\right)\right)\right)^{-\frac{1}{\kappa_{\text{c}}}} \end{aligned} \tag{8}$$

and I_0 is defined as

$$I_0\left(L_c, \kappa_c, a_{3,\min}, a_3, s\right) = \frac{1}{2 \cdot \pi^2 \cdot s^2 \cdot \frac{L_c}{a_3 \cdot (1+\kappa_c)} \cdot (a_3)^2} \cdot 1 - \frac{\cos\left(\frac{1}{\kappa_c} \cdot \arctan\left(2 \cdot \pi \cdot a_3 \cdot \frac{L_c \cdot \kappa_c}{a_3 \cdot (1+\kappa_c)} \cdot s\right)\right)}{\left(1 + 4 \cdot \pi^2 \cdot (a_3)^2 \cdot \left(\frac{L_c \cdot \kappa_c}{a_3 \cdot (1+\kappa_c)}\right)^2 \cdot s^2\right)^{\frac{1}{2 \cdot \kappa_c}}}. \tag{9}$$

Please note that the parameters L_c and and κ_c are not entered directly in CarbX but in the form of the shape factors μ and β defined as

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

$$\beta = \frac{a_3 \cdot (1 + \kappa_c)}{L_c \cdot \kappa_c}. \tag{11}$$

In addition, please note that expression $\frac{L_c}{a_3 \cdot (1+\kappa_c)} = \langle N \rangle$ is given by $\frac{\mu}{\beta}$. Furthermore, please note that $a_{3,\min}$ is given by $a_3 - \Delta_{a_3}$. a_3 and Δ_{a_3} are entered directly in CarbX.

1.3 Intralayer scattering

The intralayer scattering I_{intra} is given by

$$\begin{aligned} & I_{\text{intra}}\left(L_a, \kappa_a, l_{cc}, \sigma_1, h, k, q, s\right) \\ = & \frac{1}{n_0 \cdot S_0\left(l_{cc}\right)} \cdot \sum_{hk} m_{hk} \cdot |F_{hk}^2| \cdot J_{hk}\left(L_a, \kappa_a, l_{cc}, \sigma_1, h, k, s\right) \\ & \cdot g\left(q, l_{cc}, h, k, s\right) \end{aligned} \tag{12}$$

where h & k are Miller indices, m_{hk} is the multiplicity of the index (hk), $|F_{hk}|$ is the structure factor for a 2D hexagonal lattice, J_{hk} the normalized profile of an (hk) reflection and g describing the preferred orientation of the stacks (Faber, 2014).

The multiplicity m_{hk} of the index (hk) is given by

$$m_{00} = 1; m_{h0} = m_{hh} = 6; m_{hk} = 12 \tag{13}$$

i.e. for $h = k$ or $k = 0$ m_{hk} is 6 except for $h = k = 0$ where m_{hk} is 1; in all other cases m_{hk} is 12 (Faber, 2014).

The structure factor for a 2D hexagonal lattice $|F_{hk}|$ is given by

$$|F_{hk}| = 2 \text{ if } (h - k) = 3 \cdot n, n \in \mathbb{Z} \text{ else } |F_{hk}| = 1 \text{ (Faber, 2014)}. \tag{14}$$

The normalized profile of an (hk) reflection J_{hk} is calculated utilizing the analytical expression given in Appendix A of (Ruland and Smarsly, 2002):

$$\begin{aligned} & J_{hk}\left(L_{\mathrm{a}}, \kappa_{\mathrm{a}}, l_{\mathrm{lcc}}, \sigma_1, h, k, s\right) \\ & = \frac{1}{1/s \cdot \kappa_{\mathrm{a}}} \cdot \ \mathrm{Im}\left[\sum_{m=0}^{1/\kappa_{\mathrm{a}}-1} \frac{1/\kappa_{\mathrm{a}} - m}{m!} \cdot F\left(m, L_{\mathrm{a}}, \kappa_{\mathrm{a}}, l_{\mathrm{cc}}, \sigma_1, h, k, s\right)\right] \end{aligned} \tag{15}$$

where F is defined as

$$\begin{aligned} F\left(m, L_{\mathrm{a}}, \kappa_{\mathrm{a}}, l_{\mathrm{cc}}, \sigma_1, h, k, s\right) = & \frac{m! \cdot \left(\frac{1+\kappa_{\mathrm{a}}}{L_{\mathrm{a}} \cdot \kappa_{\mathrm{a}}}\right)^m}{\left(\frac{1+\kappa_{\mathrm{a}}}{L_{\mathrm{a}} \cdot \kappa_{\mathrm{a}}} + \frac{4 \cdot \pi^2 \cdot s_{hk}(h,k,l_{\mathrm{cc}})^2 \cdot \sigma_1^2}{3 \cdot l_{\mathrm{cc}}} - 2 \cdot \pi \cdot i \cdot s\right)^{m+1}} \\ & \cdot {}_2F_1\left[\frac{m+1}{2}, \frac{m+2}{2}, 1, \frac{-4 \cdot \pi^2 \cdot s_{hk}\left(h,k,l_{\mathrm{cc}}\right)^2}{\left(\frac{1+\kappa_{\mathrm{a}}}{L_{\mathrm{a}} \cdot \kappa_{\mathrm{a}}} + \frac{4 \cdot \pi^2 \cdot s_{hk}(h,k,l_{\mathrm{cc}})^2 \cdot \sigma_1^2}{3 \cdot l_{\mathrm{cc}}} - 2 \cdot \pi \cdot i \cdot s\right)^2}\right] \end{aligned} \tag{16}$$

and s_{hk} is defined as

$$s_{hk}\left(h,k,l_{\mathrm{cc}}\right) = \frac{\sqrt{h^2 + k^2 + h \cdot k} \cdot 2}{3 \cdot l_{\mathrm{cc}}}. \tag{17}$$

Please note that the parameters L_{a} and and κ_{a} are not entered directly in CarbX but in the form of the shape factors ν and α defined as

$$\nu = \frac{1}{\kappa_{\mathrm{a}}} \text{ and} \tag{18}$$

$$\alpha = \frac{1+\kappa_{\mathrm{a}}}{L_{\mathrm{a}} \cdot \kappa_{\mathrm{a}}}. \tag{19}$$

g, describing the preferred orientation of the stacks, is given by

$$\begin{aligned} g\left(q, l_{\mathrm{cc}}, h, k, s\right) = 1 \text{ if } q = 0; & = \frac{\sqrt{q}}{\mathrm{arctanh}\left(\sqrt{q}\right) \cdot (1+q)} \text{ if} s < s_{hk}\left(h,k,l_{\mathrm{cc}}\right) \\ & = \frac{(1+q) \cdot \sqrt{q}}{\mathrm{arctanh}\left(\sqrt{q}\right) \cdot \left[(1-q)^2 + \frac{4 \cdot q \cdot s_{hk}(h,k,l_{\mathrm{cc}})^2}{s^2}\right]} \text{ if} s \geq s_{hk}\left(h,k,l_{\mathrm{cc}}\right) \end{aligned} \tag{20}$$

(Ruland and Smarsly, 2002) & (Faber, 2014).

1.4 Incoherent Scattering

The factors for incoherent scattering are given by

$$recoil = \left(\frac{\lambda}{\lambda + 0.485 \cdot \frac{\lambda^2}{4} \cdot s^2} \right)^3 \tag{21}$$

(Ruland 1961).

$$Q_{\text{abs}} = \frac{1}{1 + 0.0485 \cdot \lambda \cdot \frac{3}{8} \cdot s^2} \tag{22}$$

(Ruland 1961).

The correction term Q is given by

$$Q = \frac{1}{y \cdot z} \tag{23}$$

where y and z are defined as follows:

$$y = 1 + \frac{\frac{\lambda^2 \cdot s}{137} \cdot \left(\frac{3.05 \cdot s^2}{0.53^2 + s^2} \right)}{b} \tag{24}$$

$$z = 1 + \frac{\pi^2 \cdot \left(\left(\frac{0.0485 \cdot s^2}{4} - \frac{1.5 \cdot 10^{-3} \cdot s^4}{(0.53^2 + s^2)^2} \right) \cdot \lambda^2 \right)^2}{\left(\frac{\lambda^2 \cdot s}{137} \cdot \left(\frac{3.05 \cdot s^2}{0.53^2 + s^2} \right) + b \right)^2} \tag{25}$$

In the equations for y and z b is the pass-band of the secondary-monochromator (if used) in Å (Ruland, 1964). The constants are given in atomic units. As the factor Q should only be used when a secondary monochromator is applied, CarbX allows the user to select whether this correction should be applied.

1.5 Correction / Tuning Factors for the Fit Curve

The correction / tuning factors are defined as follows:

$$AutoColl = \frac{r \cdot \frac{\lambda \cdot s}{2} \cdot \sin(\delta)}{l \cdot \left(\left(\frac{\lambda \cdot s}{2} \right)^2 - \sin\left(\frac{\delta}{2} \right)^2 \right)} \tag{26}$$

where r is the radius of the goniometer in centimetres, δ the divergence angle in degrees and l the irradiated length in centimetres.

$$gFact = \exp(g \cdot s) \tag{27}$$

where g is the correction value to be used and must not be confused with the function describing preferred orientation defined above. This factor corrects possibly the influence of an SAXS intensity contribution affecting the

slope of the theoretical WAXS curve at low s-values. However, it should be understood as an empirical correction factor as the SAXS intensity contribution is in general not quantifiable due to the lack of absolute intensity measurements.

$$A = 1 - \exp\left(\frac{-2 \cdot d \cdot \mu_{\mathrm{ab}}}{\sin(\theta)}\right) \tag{28}$$

where d is the thickness of the sample and μ_{ab} the absorption factor.

$$P = \frac{1 + \left(\cos\left(2 \cdot \arcsin\left(\lambda \cdot \frac{s}{2}\right)\right)\right)^2}{2}. \tag{29}$$

1.6 Data Correction Terms

The data correction terms are defined as follows:

The terms used for the correction of the position of the reflexes due to absorption are "$+Dz$" and "$D2thtx$". The numerical result of the term "$+Dz$" is added to the corresponding 2θ-value in degrees. The numerical result of the term "$D2thtx$" replaces the corresponding modulus of the scattering vector in $1/\text{Å}$, which is calculated from the corresponding 2θ-value. As these terms are implemented for special cases they should not be used generally. They are defined as follows:

$$+Dz = \frac{\sin(2\theta) \cdot \left(1 - \frac{2 \cdot d \cdot \mu_{\mathrm{ab}}}{\sin(\theta) \cdot \left(\exp\left(\frac{2 \cdot d \cdot \mu_{\mathrm{ab}}}{\sin(\theta)}\right) - 1\right)}\right) \cdot 180}{2 \cdot r \cdot \mu_{\mathrm{ab}} \cdot \pi}. \tag{30}$$

$$D2thtx = \frac{2 \cdot \sin\left(\frac{(10 + 0.25 \cdot (i-1)) + x}{2}\right)}{\lambda} \tag{31}$$

where i is the index of the data point for which the position is calculated and x defined as follows:

$$x = \frac{180 \cdot \cos(c) \cdot (d - d \cdot \coth(d \cdot \csc(c)) + \sin(c))}{\mu_{\mathrm{ab}} \cdot \pi \cdot R} \tag{32}$$

where R is a parameter for correction of positions according to D2thtx and c and f are defined as follows:

$$c = \frac{10 + 0.25 \cdot (i-1)}{2} \cdot \frac{\pi}{180} \tag{33}$$

where i is the index of the data point for which the position is calculated.

$$f = \mu_{\mathrm{ab}} \cdot t \tag{34}$$

where t is a parameter for correction of positions according to D2thtx.

The parameters used to take into account absorption by the sample are *lb* (Lambert-Beer absorption) or *pa* for a "pragmatic" approach:

$$lb = \exp\left(\frac{2 \cdot d \cdot \mu_{\mathrm{ab}}}{\sin(\theta)}\right). \tag{35}$$

$$pa = \exp\left(-1 \cdot p \cdot (2 \cdot \theta)^2\right) \tag{36}$$

where p is the correction value to be used. This term allows for a simple and pragmatic description of absorption, taking into account that an exact quantitative treatment of the absorption for the specific measurement setup can be ambiguous. It should only be used if the term *lb* defined in Eq. 35 does not provide the desired correction for absorption. The dimensionless parameter p used in this approach is presumed to lie within the interval $[10^{-5}, 10^{-3}]$. However, values above about $2 \cdot 10^{-4}$ manipulate the measurement data severely. The default value is set to $4 \cdot 10^{-5}$, a value typically used.

As the terms used in the correction for the absorption by the sample and the absorption factor defined in Eq. 28 have the same effect[1], only one of these options can be active at the same time.

2 Extensive Technical Details of CarbX'

2.1 CarbX' files and classes

In the program-wide used header files own data types and enumerations are defined. The classes for data storage are utilized to to store both data and settings such as parameter values, calculated points for the curves displayed, the autofit sequence currently used, programme states and preferences. Using and thus accessing the "Wolfram Mathematica Kernel" (provided by Wolfram) is realized by the classes for automatic fitting. A "main"-class, containing the code for initial start-up and translation and a class containing some auxiliary methods complete the programme.

[1]The absorption factor A reduces the theoretical observable intensity, the factor *lb* increases the measured intensity to correct the absorption by the sample. Activating both factors at the same time would result in double correction of absorption by the sample.

2.2 Calculation routines

In this class, each function required to calculate and manipulate the fit curve as well as each function required for corrections of measurement data is implemented as a C++ function. While only few functions are actually called from outside the class (e.g. the function providing the theoretical intensity for a given modulus of the scattering vector with the given parameters), most of the functions in the class call another function of the class, except some functions providing the basis for the calculations. These functions include interpolation functions for the atomic form factors, the different factors to be considered for Compton scattering and the basis functions for calculating the interlayer and intralayer scattering intensity. The structure can be compared to recursive algorithms regarding computational speed, although no function calls itself. As in the model, there are three blocks of functions. The first calculates the incoherent scattering. The second and third both describe the coherent scattering. The second refers to the calculation of the intralayer scattering, the third to the interlayer scattering. The rest of the functions are functions combining the results of each block, functions that correct the measurement data and some auxiliary functions as well as functions to make the class work as an object in terms of object oriented programming.

2.3 GUI

2.3.1 Main Window

The menu bar in section one contains three menus. The first menu “File” has the standard functions New, Open, Save, Save as and Exit. The second point gives access to some settings which can be set in the menu itself and to the possibility to open an extra preferences dialogue. The third point allows the user to choose a template and to reset the parameter values to the template values if a template was chosen. A template is a file containing parameter values as well as X-Y-data of a fit of a certain sample. The X-Y-data is used to display the template. The parameter values can be used as initial values for fitting data of a similar sample. The files loaded/saved using the “File” menu are NGC-files (*.ngc) which contain all necessary values to rebuild a specific fit such as the values of the sliders (see below), the corrections chosen, the wavelength and the options for the plots in both windows.

The second section contains some buttons that allow the user to open a data file, to show and hide the data and the fit curve as well as the deviation between data and fit curve, to start an attempt to automatically fit the data, to save a list of the current parameters as text file and to export the plot and/or single curves in various formats. Beside the parameters

that can be changed in the programme directly, this exported parameter list contains some parameters which are calculated from the parameters entered (see above).

The third section contains sliders for manipulating the parameters affecting the fit curve and correction of the raw data. Each value can be changed either by moving the slider or directly entering the value below the slider. Furthermore the limits for each value can be set as well as the value which should be added to or subtracted from the parameter value when the plus or minus buttons are pressed. As some parameters must not exceed certain limits, a warning is shown when this condition is violated.

The fourth section contains the plotting widget in which the measurement data and the fit curve are shown. The interval of both axes can be changed manually. However, it is also possible to enable/disable the automatic scaling of the Y-axis. Moreover, a thinning factor x can be set, which means that only each x. point is calculated.

Finally, in the fifth section, the options for the fit curve and correction of the raw data as well as the wavelength used can be set. For this purpose, a list of commonly used anode materials and the corresponding wavelengths is provided. The list can be modified by the user and is saved permanently in a separate file without overwriting the programme's default list. Each change affecting the data and/or the fit curve in any of the sections updates the corresponding curve instantly, unless deactivated in the options menu. As for some correction terms and options of the fit curve sample and the measurement equipment parameters have to be known, these parameters can also be entered in this section.

2.3.2 Supporting window to display the deviation

The deviation is calculated as

$$I_{\mathrm{dev}} = \frac{I_{\mathrm{experimental}} - I_{\mathrm{obs}}}{I_{\mathrm{obs}}} \tag{37}$$

for each point of the measured data and then plotted against the modulus of the scattering vector. Thus, the X axis of the deviation plot is scaled in the same way as the X axis of the data/fit plot in the main window. Two checkboxes allow choosing whether an interpolation of the data points of the deviation is to be shown and whether the deviation should be updated instantly after the data/fit plot in the main window has been updated. As each update consumes a few seconds of calculation time, this option is disabled by default and the plot has to be updated manually by using the "update plot" button. Export of the deviation data either as an image or a text file is possible by the corresponding buttons, as well as the closing of the window.

2.3.3 Supporting window for automatic fitting

The commands for an automatic fitting procedure, named here "autofit sequence", are stored in an AFS-file (*.afs) in the subfolder "data/autofitsequences" relative to the "CarbX.exe" file for each autofit sequence created by the user. A default autofit sequence is supplied in this folder and loaded by default each time the button "automatic fitting" in the main window is clicked. An autofit sequence consists of an arbitrary number of autofit steps. For each step the parameters to be fitted and the respective limits (minimum and maximum) for these parameters are defined together with possibly used additional conditions concerning two or more parameters. The name of the autofit sequence, from which the file name for the AFS-file is derived, the name of each step and which steps are to be used are stored in addition.

For the manipulation of the autofit sequences the supporting window (Fig S10) contains several elements. At the left bottom, there are buttons for choosing, adding, saving, renaming and deleting a autofit sequence. Choosing a sequence discards the changes made (a warning is displayed, see below), takes over the values from the main window again and updates the elements used to display the values of the sequence. When adding a sequence, a new sequence with the name entered by the user after a corresponding prompt and the values of the sequence selected before is created and the corresponding AFS-file is stored on the hard disk. Deleting a sequence also deletes the corresponding AFS-file. It is possible to delete all sequences, which, however, disables all functions except the function to add the current sequence again, as the AFS-files are in general only altered when explicitly requested. Otherwise, all changes are stored non-permanently in the memory. In addition, the current sequence is set to a default sequence consisting of one step with all parameters set for fitting. When renaming a sequence the user is asked to enter the new name. This name is only stored on the hard disk if the sequence is saved afterwards. In general, the user can only choose names for sequences on adding or renaming that do not exist already or are the same as the name before renaming. Due to restrictions within the file system of Windows, names that contain different illegal characters for filenames at the same position may be recognized as the same name.

The steps of the autofit sequence chosen are listed in the bottom middle with a green background for an active step and a red background for a inactive step. Selecting a step updates the elements above and on the right of the list to manipulate the step. Each step can be shifted up or down in the sequence (the steps are executed in the listed order), be renamed, be deleted, disabled / enabled and executed as a single step even if disabled. Since at least one active step must be present, it is not possible to delete or disable all steps.

If the last active step is deleted, the step selected after deletion is enabled automatically. For each step, the user can add additional conditions as well as alter and delete existing additional conditions. The conditions entered are not validated. Furthermore, the user can select which parameters are to be fitted in the step currently selected by activating / deactivating the corresponding checkboxes. If a checkbox is activated, the user can also enter the limits for automatic fitting or the character 'n' to disable the limit(s) for the associated parameter. To restore the value used before disabling the border, the character 'u' has to be typed. Alternatively, a numeric value can be entered. Other inputs are ignored and the input field is set back to the former value.

If changes made to an autofit sequence are to be discarded either by selecting another sequence or by closing the window, the user is asked whether the changes are to be saved. The initial values for automatic fitting, respectively the values for the parameters not being fitted in the step currently selected are also displayed near to the checkboxes for the corresponding parameter. The user can reset the value to the value used in the main window either for an single parameter or for all parameters using the corresponding buttons. The latter disables the possibility to preview and take over the results of automatic fitting. If the initial value for automatic fitting is smaller than the minimum or larger than the maximum, the corresponding field is highlighted in yellow and the starting value is automatically set to the minimum or maximum during automatic fitting. The fields also display the parameters obtained by automatic fitting, after the sequence or a single step is executed by clicking the corresponding button. Both for a single step and the whole sequence the fit program displayed below the button to execute the whole autofitting sequence is used. Currently, only Wolfram Mathematica is supported.

To reduce the time for automatic fitting, the user can set a data thinning factor, so that only each x. point of the measurement data is used for automatic fitting. After executing a single step or the whole sequence, the results of automatic fitting can be displayed temporarily in the main and deviation windows or taken over to the main window. The latter also closes the supporting window for automatic fitting. If automatic fitting has been performed and the supporting window is to be closed by other user inputs, the user is asked whether the results shall be taken over into the main window.

After activating the supporting window for automatic fitting, several input elements in the main window as well as the possibility to close the main window are disabled. They are re-enabled after closing supporting window, as in general only the tuning of the parameter values and the settings options for the display of the plot are allowed. Further input elements are disabled

when the results of the automatic fitting are previewed, as changes made while the results are displayed would have no effect. If the parameter g is disabled in the main window, the corresponding input elements are hidden in the supporting window for automatic fitting, thus the settings for this parameter cannot be modified. Furthermore, the parameter is neglected in automatic fitting, regardless of the values stored in the AFS-file.

2.3.4 Colour code used in the main window and the supporting window for automatic fitting

The input elements for the parameters, the options for the fit curve and the data correction used in both the main window and the supporting window for automatic fitting are coloured for easy association according to Table 1.

Table 1: Overview of the parameter groups, their colours and meaning

parameter group	meaning
I_{inter}	parameters for coherent interlayer scattering
I_{intra}	parameters for coherent intralayer scattering
I_{coh}	parameters for coherent intralayer in general
I_{inc}	parameters for incoherent (Compton) scattering
$corr_{\text{fit}}$	parameters for correction of the fit curve
$corr_{\text{data}}$	parameters for the correction of the measured values
misc	various parameters (wavelength / absorption factor) for multiple sections
other	other parameters

2.3.5 Special dialogues

The dialogue for setting the format of the raw data consists only of two combination boxes allowing the user to select the right format and one button to confirm the chosen formats.

The dialogue to set the programme options allows defining a default directory which is opened in all following file dialogues and to choose whether the full path of files or only the file name is to be shown in the window and button captions. Furthermore it allows choosing if the interval of the X axis of the data/fit plot is to be reset after loading another fit-file or using the "File/New" command. In addition, the thickness of curve lines and curve dots can be adjusted and the element in front can be selected. Moreover, the the template dialogue after loading a data file and the automatic calculation of templates can be enabled/disabled. Finally, the path to the "Wolfram Mathematica Kernel", which is necessary for the automatic fitting, can be

stored. To perform automatic fitting, the path of the "MathKernel.exe" located in the installation directory of "Wolfram Mathematica" has to be entered in the dialogue described here. Any "Wolfram Mathematica" version supporting "WSTP" version 4 can be used.

The template dialogue, shown after loading a data file if enabled, displays eight plotting widgets containing plots of the template files placed around a central plotting widget containing the plot of the measurement data. Depending on the setting for the automatic calculation of the templates, either the X-Y-data stored in the template file is used or the points for each template are calculated. The latter also applies if a template file contains no X-Y-data. In this case the points are calculated automatically. As the calculation needs some time, it takes several seconds until the dialogue shows if the automatic calculation of the templates is enabled. After appearance of the dialogue the user can either choose the most suitable template by selecting its number in the box left of the button and clicking on this button or choose no template by clicking on the other button below both elements mentioned before. Furthermore, the user can request to calculate the points for all templates as long as the calculations have not been executed yet. This means the corresponding button is only available if X-Y-data have been read and the request for calculation has not been made yet. If the parameters (wavelength and thickness of the sample if the correction for absorption is enabled for the specific template) used to calculate the X-Y-data differ from the current programme parameters for at least one template, the user is recommended to recalculate the templates. CarbX comes with eight templates stored in the subfolder "data/templates" relative to the "CarbX.exe" file. These files are named "# .tpl" where "# " stands for a integer number between one and eight. The files can be replaced with own template files but this folder has always to contain eight files named according to the convention mentioned before.

The dialogue for setting the name of a template consists only of a caption, a text field for entering the desired name and a button to confirm the name. The text field is prefilled with either the filename of the fit loaded (if so), the filename of the loaded measurement file or nothing. The suggested directory to save the template is not the default directory set in the programme options but the directory of the loaded measurement file or fit, respectively. The suggested or self-entered name can be confirmed either by pressing the enter key in the text field or by clicking on the corresponding button.

3 Required steps for three typical application cases of CarbX

Prior to executing the steps described below, download and extraction of the CarbX programme files is required. In addition, the preferences as well as the wavelength used for measuring have to be set. For all application cases, it is possibly the change the scale of both axis in order to examine certain segments of the fit (curve) in more detail.

3.1 Display of the theoretical curve

To display the theoretical fit curve only, a single click on the button "draw fit curve" (Fig S3) is sufficient. However, depending on the calculation power of the computer used, it should be considered to set a reasonable thinning-factor by using the spinbox shown in Fig S4 and possibly the settings for the fit curve as well as reasonable (start)values for the parameters before displaying the fit curve. Alternatively, the automatic update of the fit can be deactivated and manual updating be utilized. In general, direct editing of the parameter values is less time consuming than using the sliders (see Fig S7) to change the value. Finally, the current work can be saved as a NGC-file.

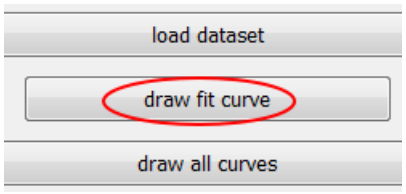

Figure S3: Screenshot of the button "draw fit curve" with its caption red encircled.

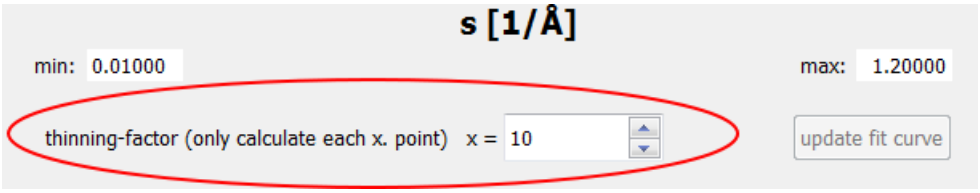

Figure S4: Screenshot of the red encircled spinbox to set the thinning factor.

3.2 Manual fitting of data

For manual fitting of data, is required to load the desired data using the button "load dataset" (Fig S5) and subsequently set the correct format of the raw data in the corresponding dialogue (Fig S6). The data is then displayed automatically. It is recommended to save the current work in a meaningful named NGC-file. Next, setting of the desired options for data correction and the fit curve, a reasonable data thinning factor by using the spinbox shown in Fig S4 and coarse start values for the parameters using the element displayed in Fig S7 is recommended. At this stage one can open the dialogue to choose a template (Fig S8). If a template appears suitable, it is recommended to choose it as it is very likely to ease the the manual fitting procedure. However, neither opening the template dialogue, nor choosing are template are mandatory. Subsequently, the theoretical curve can be displayed by utilizing the button "draw fit curve" (Fig S3). Now, manual adapting of the fit parameters is required until a satisfactory fit is obtained. It is advisable to first refine the underground (k, $const_1$ and $const_2$), second the peak positions (a_3, l_{cc}) and third the peak shape (μ, β & σ_3 and (ν), α & σ_1) before changing other parameters. In this course ν should be set to and kept at a fixed value. If automatic fitting is to be executed, a coarse fit is sufficient. For computers with limited calculation power, it is recommend to enter the parameters values directly instead of using the sliders (see Fig S7). Finally, the results can then be analysed regarding to deviation between data and fit and be exported in several formats.

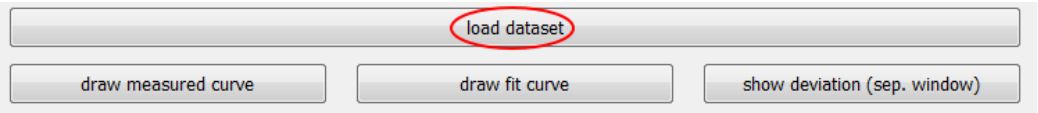

Figure S5: Screenshot of the button "load dataset" with its caption red encircled.

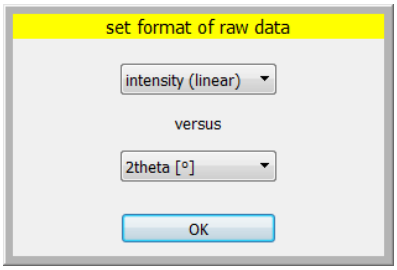

Figure S6: Screenshot of the dialogue for for setting the format of the raw data.

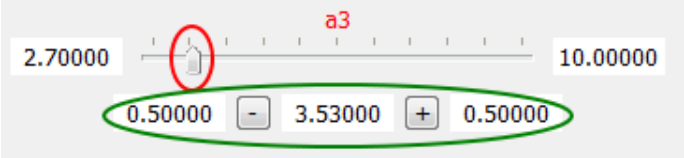

Figure S7: Example of a slider structure to set parameter values. The value can either be set by moving the red encircled slider or by using the green encircled elements to type in the value directly or increment/decrement it, respectively.

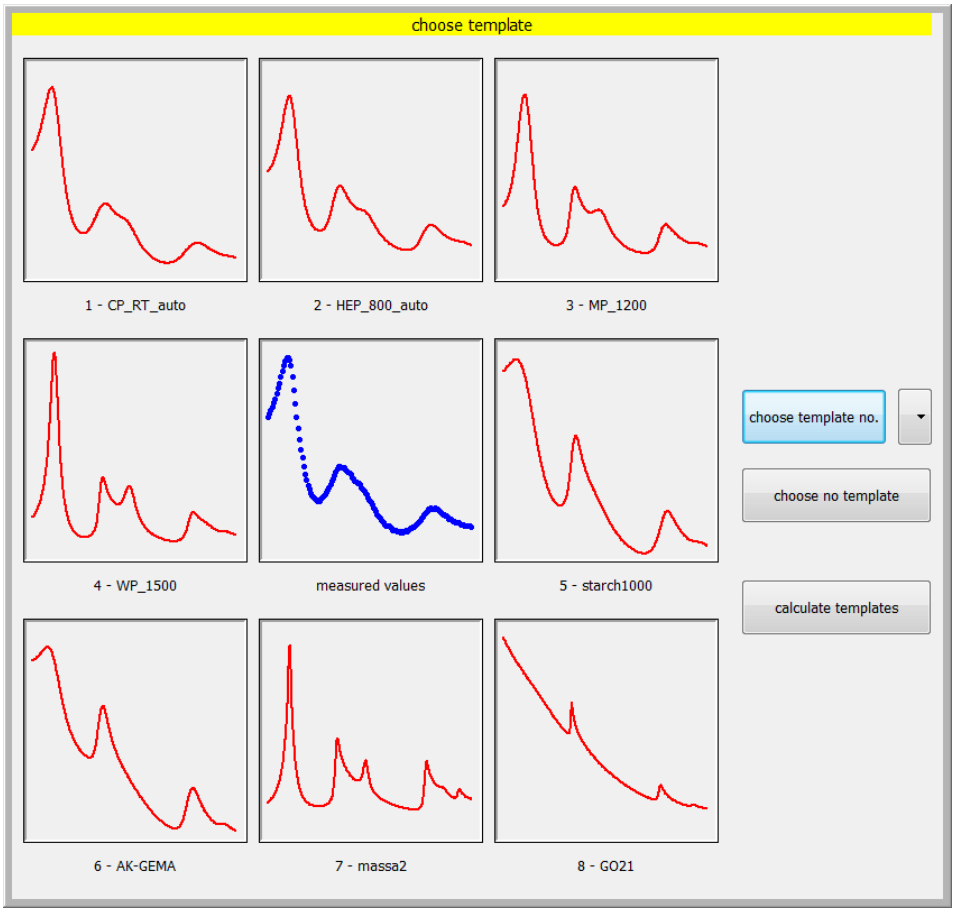

Figure S8: Screenshot of the dialogue to choose a template.

3.3 Automatic fitting of data

Prior to automatic fitting of data, a coarse manual fit is to be obtained. The automatic fitting is performed using a supporting window (shown in , which can be displayed by clicking the button "automatic fitting" (Fig S9). After choosing and/or editing the desired autofit sequence, setting the data thinning factor to be used for automatic fitting and selecting the desired programme for automatic fitting (see Fig S10 and article for more details), automatic fitting can be executed. While for convenience the execution of the whole sequence at once is recommended, executing the steps one after another can be helpful to identify automatic fitting problems. Finally, after execution of automatic fitting, the results can be previewed, and if satisfactory, be taken over into the main window. The results can then be analysed regarding to deviation between data and fit (the corresponding supporting window is activated by clicking on the button displayed in Fig S11) and be exported in several formats (the corresponding buttons are shown in Fig S12.

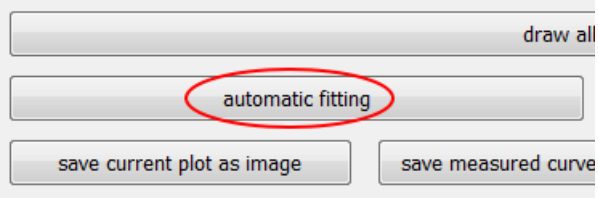

Figure S9: Screenshot of the button "automatic fitting" with its caption red encircled.

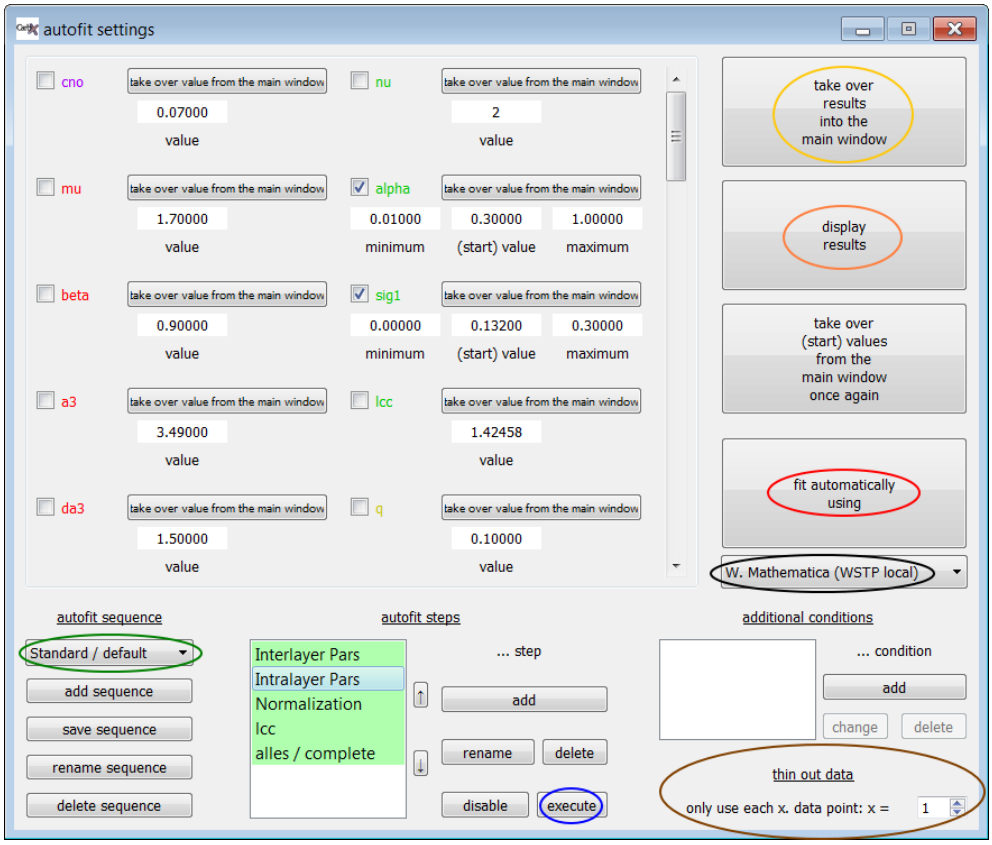

Figure S10: Screenshot of the supporting window for performing automatic fitting. The desired autofit sequence can be chosen by using the green encircled drop-down-list. The data thinning factor is set using the brown encircled spinbox. The programme for automatic is chosen by using the black-encircled drop-down-list. Automatic fitting is started by clicking on the button with the red encircled caption. After automatic fitting, previewing the results can be achieved by using the orange encircled button. For final takeover of the automatic fitting results, the yellow encircled button has to be clicked. Execution of a single automatic fitting step is possible by clicking on the button with the blue encircled caption.

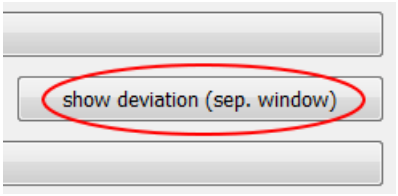

Figure S11: Screenshot of the button to display a supporting window for displaying the deviation between fit and data with its caption red encircled.

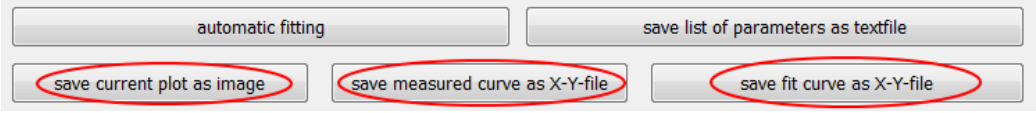

Figure S12: Screenshot of the buttons to export results of (automatic) fitting with their captions red encircled.

4 Hints for manual fitting of the sample file "HEP_RT.xy"

1. The template "HP_800" provides reliable initial values for the manual fit as depicted in Fig S13.

2. After selecting the template setting $const_2$ to 1.25 already yields a very coarse fit as depicted in Fig S14.

3. The position of the $(00l)$ reflexes can be adjusted by setting the a_3 value to 3.47. In addition, the Δ_{a_3} value should be set to the recommend maximum of 0.77 as $a_{3,\mathrm{min}}$ should be at least 0.7. The result is depicted in Fig S15.

4. The shape of the $(00l)$ reflexes can be adjusted by setting the shape factors μ and β to 1.67 and 0.9 respectively together with σ_3 to 0.2. The result is depicted in Fig S16.

5. The shape of the (hk) reflexes can be adjusted by setting the shape factors ν and α to 2 and 0.27 respectively together with σ_1 to 0.14. The result is depicted in Fig S17.

6. At this point, the influence of the preferred orientation of the stacks and the anisotropy of the atomic scattering factor for carbon perpendicular to the graphene sheets has to be correctly considered. This can be achieved by setting q to 0.03 and Δ_{an} to 25. This yields a fair manual fit as depicted in Fig S18

7. After fine tuning the factors for background consideration, achievable by setting $const_1$ to 730 and the slope of the fit curve, achievable by setting g to 0.27 a good manual fit is achieved as depicted in Fig S19. Good means that there is merely statistical deviation between data and fit as shown in Fig S20.

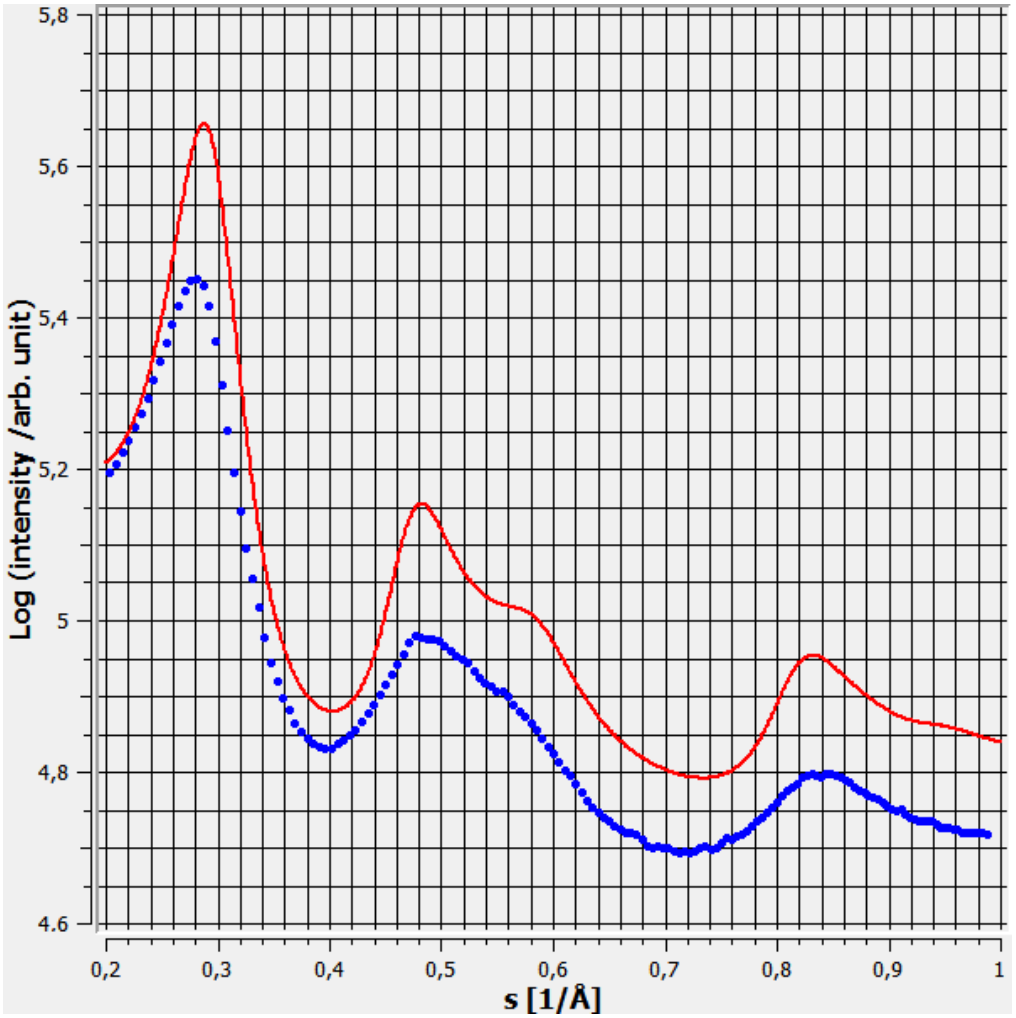

Figure S13: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after choosing the template "HP_800" is depicted.

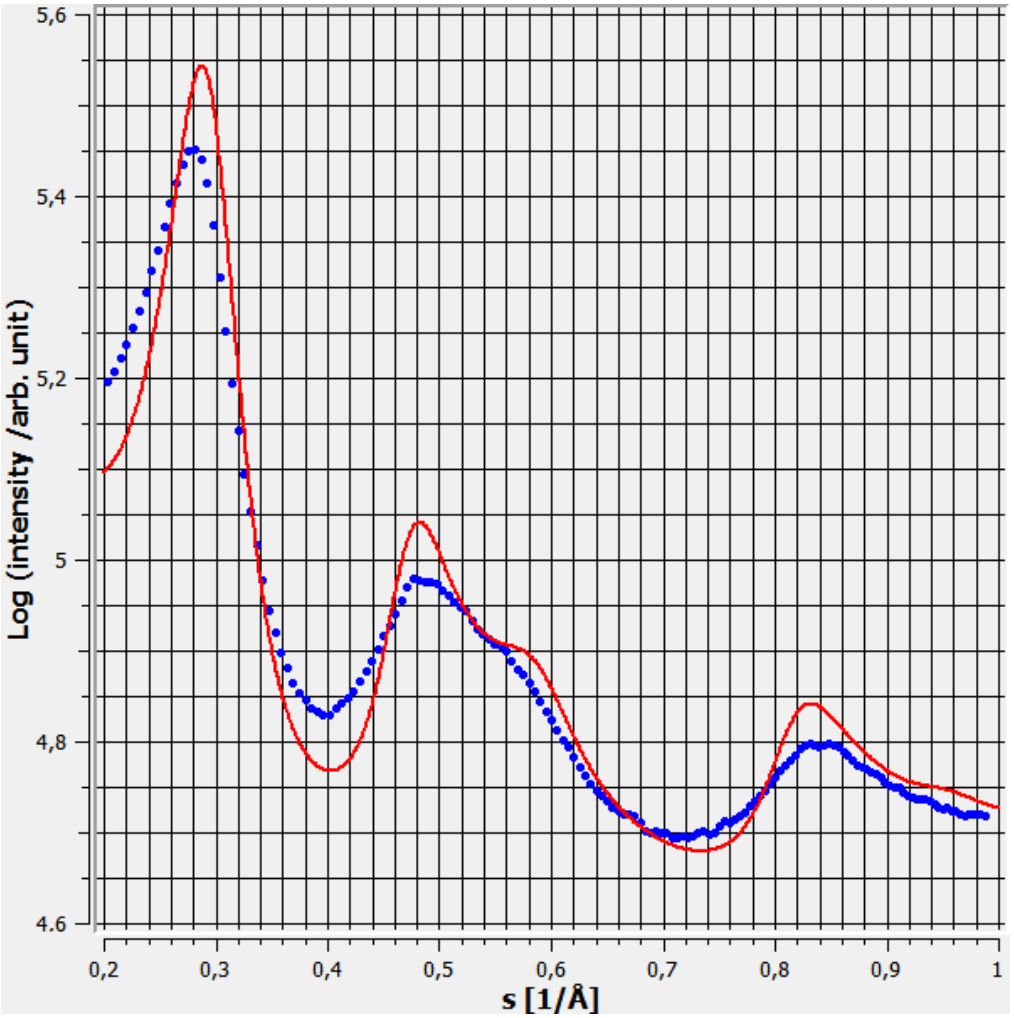

Figure S14: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after setting the factors to consider a background is depicted.

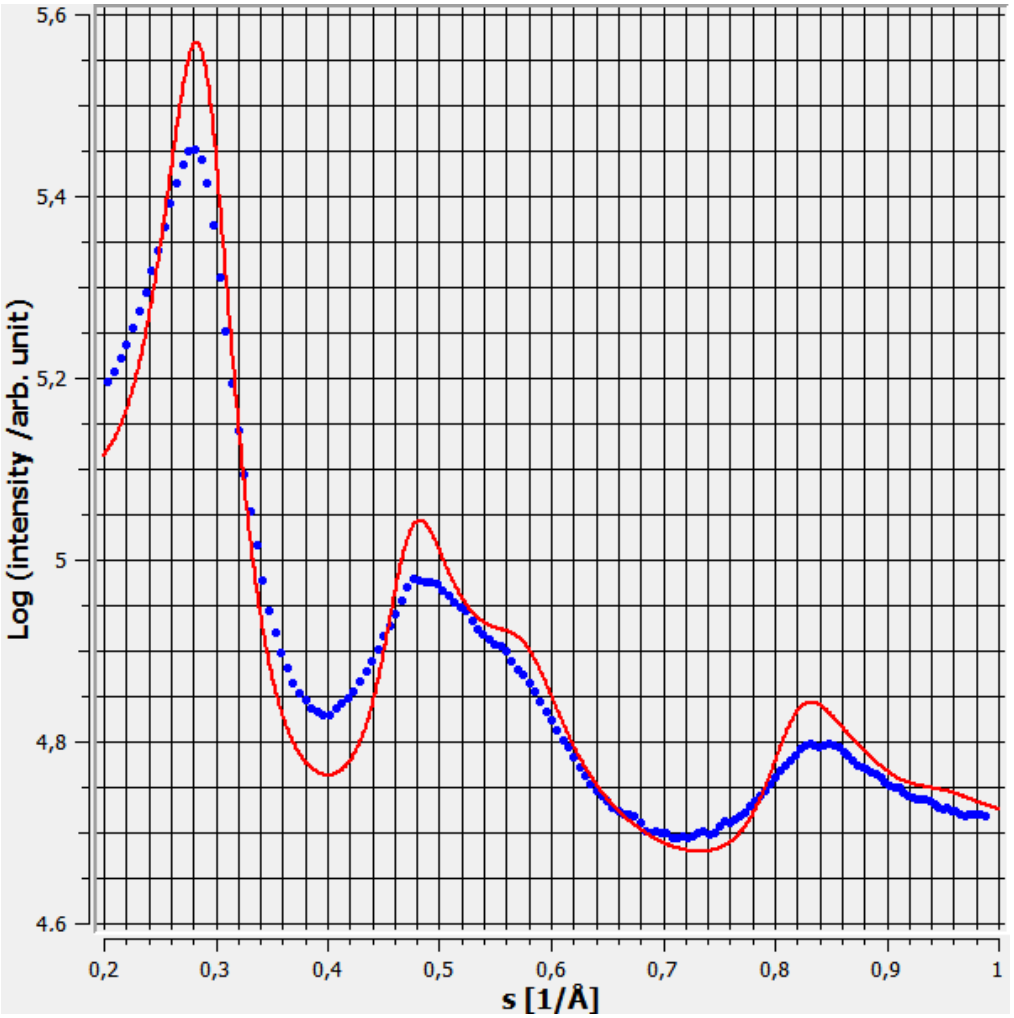

Figure S15: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after setting the parameters influencing the positions of the (00l) reflexes is depicted.

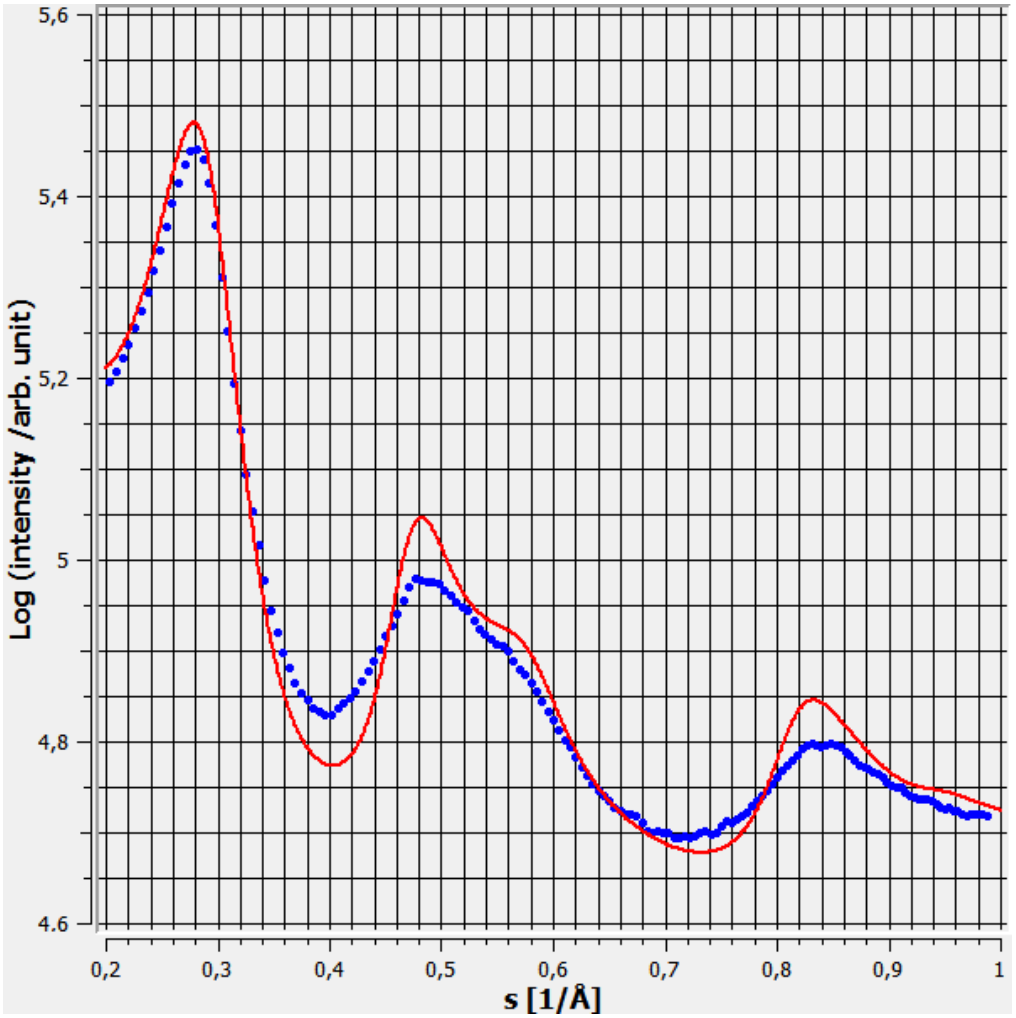

Figure S16: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after setting the parameters influencing the shape of the $(00l)$ reflexes is depicted.

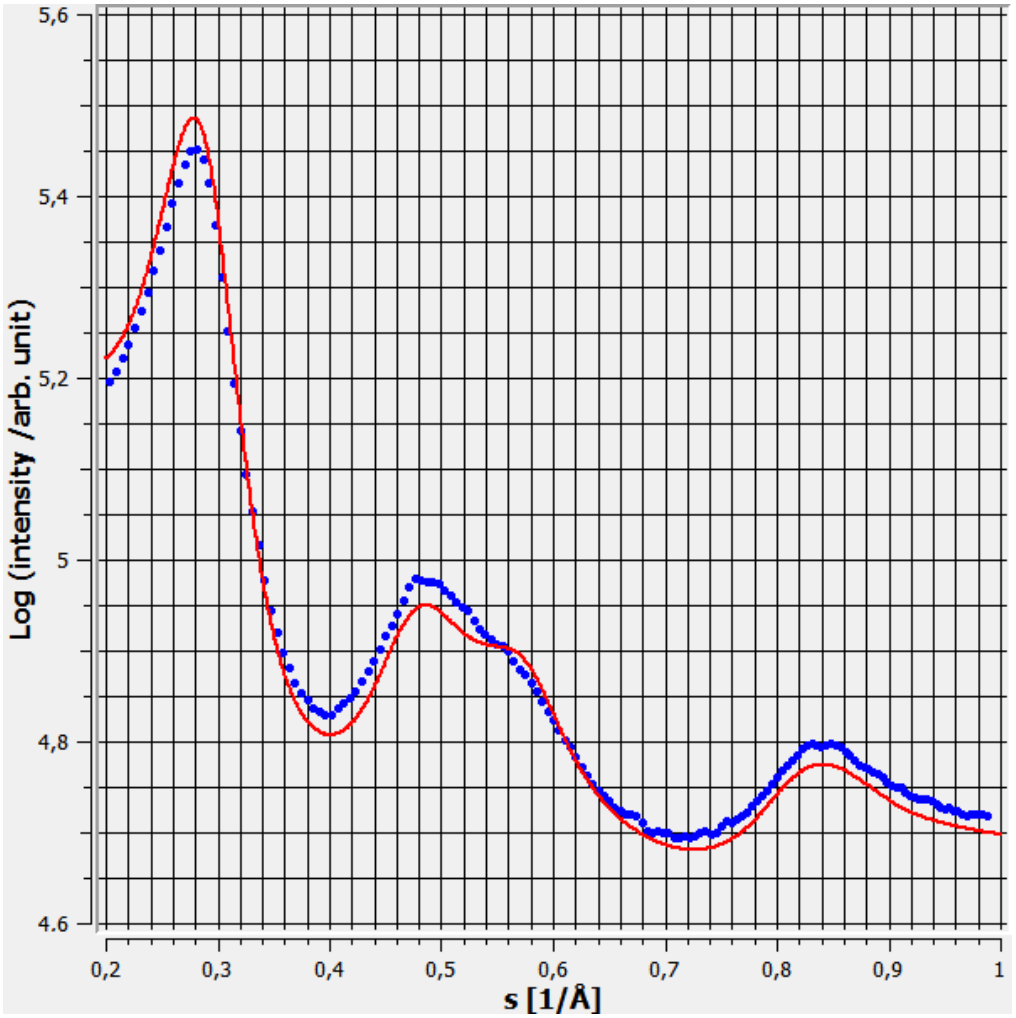

Figure S17: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after setting the parameters influencing the shape of the (hk) reflexes is depicted.

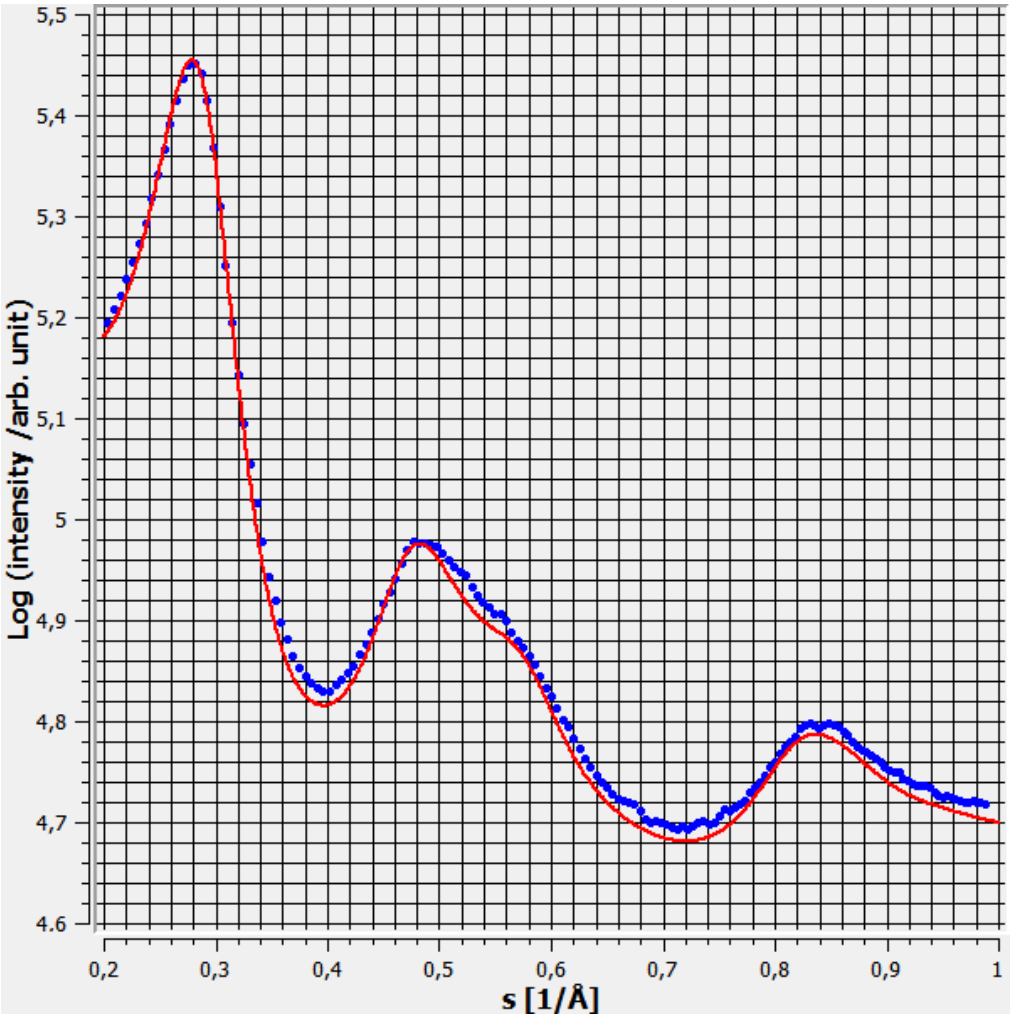

Figure S18: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after setting the parameters influencing the consideration of the preferred orientation of the stacks and the anisotropy of the atomic scattering factor for carbon perpendicular to the graphene sheets is depicted.

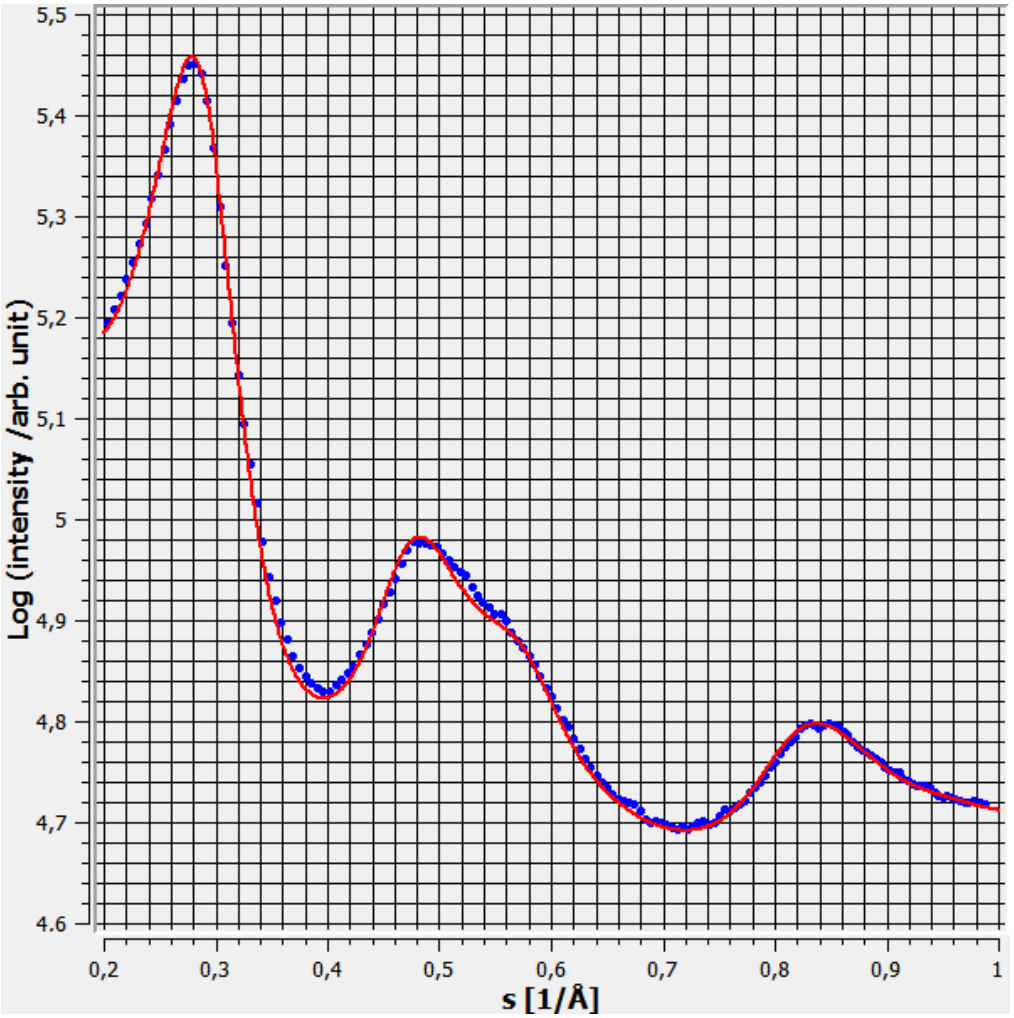

Figure S19: Fit (red solid line) of the WAXS data (blue dots) of the HEP_RT sample file. Here the state after after fine tuning the factors for background consideration and tuning the slope of the fit curve is depicted.

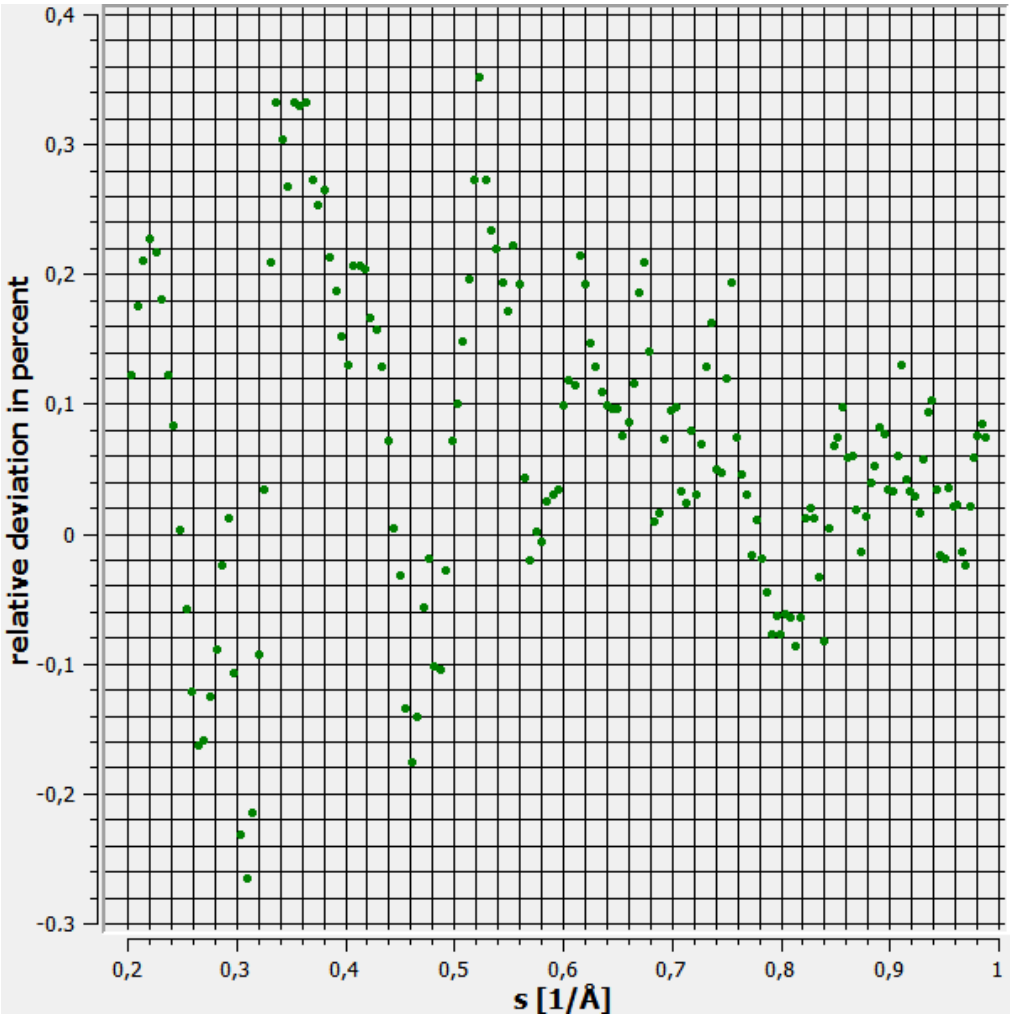

Figure S20: Relative deviation between fit and data for the the HEP_RT sample file. As merely merely statistical deviation between data and fit is present, the fit quality can be rated as “good”.

References

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