EXPERI

Users Guide

Version 6.21

written by

Reinhard Neder
Email: reinhard.neder@fau.de

Thomas Proffen
Email: tproffen@ornl.gov

http://tproffen.github.io/DiffuseCode

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Preface

Disclaimer

The EXPERI software described in this guide is provided without warranty of any kind. No liability is taken for any loss or damages, direct or indirect, that may result through the use of EXPERI. No warranty is made with respect to this manual, or the program and functions therein. There are no warranties that the programs are free of error, or that they are consistent with any standard, or that they will meet the requirement for a particular application. The programs and the manual have been thoroughly checked. Nevertheless, it can not be guaranteed that the manual is correct and up-to-date in every detail. This manual and the EXPERI program may be changed without notice.
EXPERI is intended as a public domain program. It may be used free of charge. Any commercial use is, however, not allowed without the explicit written permission of the authors.

Using EXPERI

Publication of results totally or partially obtained using the program EXPERI should state that EXPERI was used and contain the following reference:

> PROFFEN, TH. & NEDER, R.B. (1997) "DISCUS, a Program for Diffuse Scattering and Defect Structure Simulations" *J. Appl. Cryst.*, **30**, 171-175

More information

This users guide can only provide program specific details. A broader discussion of simulation techniques and many EXPERI examples and macro files can be found in our book

> NEDER, R.B. & PROFFEN, TH. "Diffuse Scattering and Defect Structure Simulations - A cook book using the programs DISCUS", *IUCr Texts on Crystallography*, Oxford University Press, 2008.

The macro files used in the cookbook can also be downloaded to the DISCUS homepage at `https://github.com/tproffen/DiscusCookBookExamples`.

Contents

Chapter 1

Introduction

1.1 What is EXPERI ?

EXPERIis the experimental data section of the DISCUS SUITE. It provides tools to handle experimental data for further processing.

1.2 Getting started

After the program DISCUS SUITE is installed properly and the environment variables are set, the program can be started by typing 'discus_suite' at the operating systems prompt for a Linux or MACos operating system. For a Windows operating system, double click on the `discus_suite` icon on your desktop.

Symbol	Description
"text"	Text given in double quotes is to be understood as typed.
<text>	Text given in angled brackets is to be replaced by an appropriate value, if the corresponding line is used in EXPERI. It could, for example be the actual name of a file, or a numerical value.
`text`	Text in single quotes exclusively refers to EXPERI commands.
[text]	Text in square brackets describes an optional parameter or command. If omitted, a default value is used, else the complete text given in the square brackets is to be typed.
{text | text}	Text given in curly brackets is a list of alternative parameters. A vertical line separates two alternative, mutually exclusive parameters.

Table 1.1: Used symbols

The program uses a command language to interact with the user. The command `exit` terminates the program and returns control to the shell. All commands of EXPERI consist of a command verb, optionally followed by one or more parameters. All parameters must be separated from one another by a comma ",". There is no predefined need for any specific sequence of commands. EXPERI is case sensitive, all commands and alphabetic parameters MUST be

typed in lower case letters. If EXPERI has been compiled using the `-DREADLINE` option (see installation files) basic line editing and recall of commands is possible. For more information refer to the reference manual or check the online help using (`help command input`). Names of input or output files are to be typed as they will be expected by the shell. If necessary include a path to the file. All commands may be abbreviated to the shortest unique possibility. At least a single space is needed between the command verb and the first parameter. No comma is to precede the first parameter. A line can be marked as comment by inserting a "#" as first character in the line.
The symbols used throughout this manual to describe commands, command parameters, or explicit text used by the program EXPERI are listed in Table 1.1. There are several sources of information, first EXPERI has a build in online help, which can be accessed by entering the command `help` or if help for a particular command <cmd> is wanted by `help <cmd>`. This manual describes background and principle functions of EXPERI and should give some insight in the ways to use this program.
EXPERI is distributed as part of the diffuse scattering simulation software DISCUS. However, EXPERI can be used as general refinement program separate from the DISCUS program package.

1.3 Command language

The program includes a FORTRAN style interpreter that allows the user to program complex modifications. A detailed discussion about the command language which is common to all DISCUS package programs can be found in the separate DISCUS package reference guide which is included with the package.

Chapter 2

Experimental data treatment

This chapter deals with the conversion of powder diffraction data to the powder pair distribution function (PDF). In short, the PDF is the calculated as a sine-Fourier transform of the powder diffraction pattern after suitable initial data processing.
An experimental powder diffraction pattern consists of several contributions. The main contribution of interest is the coherently scattered intensity signal of your sample. This will consist of the Bragg reflectons and possibly a broad diffuse scattering signal. All of the remaining signal is commonly termed the background, even though it consists of several different contributions. There will be scattering by a sample support, by air, and by any sample environment such as a furnace, pressure cells and so on. Further *background* contributions arise from incoherent scattering by the sample such as incoherent thermal diffuse scattering. In case of xray scattering a large contribution at large values of $Q = 4\pi sin(\Theta)/\lambda)$ is incoherent Compton scattering.
The conversion of an experimental powder diffraction pattern follows the following steps:

- Subtract properly scaled backgroud data, usually obtained from an *empty* measurement
- Subtract the remaining incoherent background such as thermal diffuse scattering and Cmpton scattering
- Normalize the intensity onto an absolute scale
- divide by the average atom form factor squared $< f >^2$ to obtain the normalized structure function S(Q)
- Subtract 1 from S(Q) and point wise ultiply with Q to yield the reduced normalized structure function F(Q): $F(Q) = Q[S(Q] - 1]$
- perform the sine Fourier transform to yield the commonly used form of the pair distribution function G(r): $G(r) = \frac{1}{2\pi} \int_{Q=Q_{min}}^{Q_{max}} F(Q)sin(Qr)dr$

Somewhat surprisingly, the background substraction and scaling can be performed with an empirical procedure as described by Billinge and Farrow (2013). The `exp2pdf` section within EXPERI uses an algorithm inspired by this paper.

2.1 Powder PDF

The atomic pair distribution function (PDF) can be obtained from powder diffraction data and is a valuable tools for the study of the *local* atomic arrangements in a material. This section describes how DISCUS can be used to transform an experimental powder diffraction pattern to a powder PDF.
The commonly used powder PDF is obtained from the reduced normalized intensity via a sine Fourier transform.

$$G_{obs}(r) = \frac{2}{\pi} \int_{Q_{min}}^{Q_{max}} F(Q) sin(Qr) dr \tag{2.1}$$

where F(Q) is obtained from the normalized intensity as:

$$F(Q) = Q[S(Q) - 1] \tag{2.2}$$

The normalized intensity in turn is generated from the experimental powder diffraction pattern through the application of several corrections. These corrections treat aspects like treat inelastic scattering etc.
For quite some time now, empirical corrections are used that turn out to perform a good job, see Billinge and Farrow (2013) for more detaiuled discussion. The algorithm in DISCUS essentially performs along the lines described in Billinge and Farrow (2013).

2.1.1 Treatment of powder data in DISCUS

The experimental powder pattern is transformed within DISCUS by performing the following steps:
First the input data are transformed onto an equidistant grid in Q-space. The same is done for background data, if the user provided a background measurement. The initial steps along Q for the powder pattern and the background pattern do not have to be identical. The only two requirements are that Q_{min} of the powder data is equal or lower than the corresponding limit for the powder data. And secondly that the background Q_{max} is higher that the limit for the data.
If background data are provided, these are subtracted from the powder intensities. An optional scale factor allows to take different counting times into account.

$$I_p(Q) = I_{obs} * scale \cdot Background \tag{2.3}$$

Next DISCUS calculates the average form factors squared $\langle f \rangle^2$, using the composition provided by the user or alternatively the composition of the current structure. This step is essential, as it is the only multiplicative correction that is applied. The user should ensure that the composition is correct or at least close to the actual composition.

$$I_i(Q) = \frac{I_p}{\langle f \rangle^2} \tag{2.4}$$

The result is point wise multiplied by Q to obtain an intermediate step towards F(Q).

$$F_i(Q) = Q \cdot I_i(Q) \tag{2.5}$$

The next step is the empirical transformation into the actual F(Q). A polynomial function is fitted through these intermediate data. The order of the polynomial can be adapted by the user. The difference between the intermediate data and the polynomial is taken as F(Q). As the order of the polynomial should be kep moderately low, the polynomial itself is a slowly varying function in reciprocal space. Its highest frequency is much lower than that of the actual diffraction signal by the sample.

$$F(Q) = F_i(Q) - \sum_i p_i \cdot Q^i \tag{2.6}$$

At this point the Q-scale can be adapted, which is likely necessary for electron diffraction data only. As the effective camera length might vary slightly, DISCUS allowes to determine the peak position of a significant maximmum in F(Q) and to scale the Q-axis with a multiplicative operation to set the peak position to a user provided expected value. Be aware that this scaleing operation effectively prohibits any interpretation of absolute interatomic distances. Relative lengths of different pair distances also affected by this scale.
The empirical transformation of the powder intensity onto F(Q) does not include a transformation onto an absolute scale. Instead DISCUS applies a multiplicative operation to F(Q) to put this onto an empirically determined approximate absolute scale. This the integral over a peak in G(r) does not allow you to determine an absolute coordination number. As the powder diffraction menu in DISCUS includes a multiplicative scale factor you can still fit the parameters of a model structure to obtain a match between the observed and experimental PDF.
As last step DISCUS applies Eq. 2.1 to obtain the observed powder PDF. User supplied limits allow to flexibly write different sections of the PDF.

2.1.2 Example

The macro in this example illustrates such a data transformation in DISCUS.

```
 1 exp2pdf
 2 reset
 3 data xy, DATA/powder.inte
 4 back xy, DATA/background.inte, scale:1.0
 5 radiation xray
 6 comp comp:ZnO
 7 limits inst:[,30.0], fourier:[29.5]
 8 poly order:11
 9 qscale qobs:2.00, qcrystal:2.001
10 output gr:GROBS/sample.grobs, rmin:0.01, rmax:100.00, rstep:0.01
11 output iq:GROBS/sample.iqobs
12 output sq:GROBS/sample.sqobs
13 output fq:GROBS/sample.fqobs
14 run mode:inter
15 exit  ! Back to the main DISCUS menu
```

The DISCUS command `exp2pdf` steps into the data treatment menu. As for all menus in DISCUS the `reset` command (line 2) allows you to ensure that all parameters are reset to their initial values at program start.

In lines 3 and 4 the experimental data and the background are read from the corresponding input files. DISCUS can handle several input formats like simples 2 or 4 column ASCII files, Spec type files or generic CSV type files. See the on-line help in KUPLOT for the `load` command for further details.
The empirical algorithm in DISCUS can be applied to `xray`, `neutron` or `electron` diffraction data, simply choose the appropriate value on the `radiation` command (line 5).
The composition is set in line 6 with `comp` command. You can specify the actiul composition either with the optional parameter `comp:` followed by a chemical statement or likewise just as a simple parameter on the `comp` command line. The composition should be specified as a list of atom names, optionally followed by the (relative) abundance. At names must have a capital first letter and a lower case second letter. Spaces are irrelevant. Internally DISCUS normalizes the sum of all element abundances to 1. Thus the following lines would give exactly the same composition:

```
comp comp:Zno
comp comp:Zn1.0O1.0
comp comp:Zn 1.0 O 1.0
comp Zn 2 O 2.0
```

Alternatively you can use the current structure with in DISCUS to specify the composition, in this case the command should be:

```
comp comp:current
```

As data at high Q values might be affected by high noise level and detector artefacts, DISCUS allows yo to limit the upper Q-value to an `instrumental` value on the `limits` command line. Data above this Q-value will be ignored and have no effect on the calculations. Especially for electron diffraction, the low Q data may likewise by overshadowed by the primary beam. In this case a lower limit can be applied as well. The following lines illustrate the options for the `inst:` parameter:

```
inst:[qmin_i, qmax_i]   ! User supplied lower and upper instrumental limit
inst:[qmin_i,       ]   ! Lower limit only, upper defaults to Qmax in data set
inst:         qmax_i    ! Upper limit only, lower defaults to Qmin in data set
inst:[        qmax_i]   ! Upper limit only, lower defaults to Qmin in data set
inst:[      , qmax_i]   ! Upper limit only, lower defaults to Qmin in data set
```

To avoid unnecessary Fourier termination ripples, the sine-Fourier transform of Eq. 2.1 should be limited at a Q_{max} where F(Q_{max}) is zero. DISCUS will determine such a Q_{max} value either below $Q_{max;inst}$ or close to a user supplied value on the `fourier:` parameter.
The lower limit Q_{min} in the integeral, Eq. 2.1 usually is less critical. Most of the times DISCUS can determine this value automatically as the first significant minimum in F(Q). Data below Q_{min} are replaced by a straight line to the point Q=0; F(0) = 0. If necessary include a Q_{min} value on the `fourier:` paarameter.

```
fourier:[qmin_four, qmax_four]
```

The default polynomial extends to 9the order. An indication that this order is too low is a peak in the experimental G(r) at very short distance below 0.5Å. If necessary try to expand the order. As a further test, run the following macro within KUPLOT immediately after the `exp2pdf` macro is finished.

```
fit n[1]
show
exit
```

The transformation creates as last KUPLOT data set F(Q). The macro will list the parameter values and their estimated uncertainty. Ensure that the highest order are still significant.
The observed PDF is always written into a default file name. Most of the times though the user will choose a suitable file name and suitable limits for the distance r on a `output` command line, line 10. In the example output data will be witten into the directory `GROBS`, file `sample.grobs`. The distance values on teh optional parameters `rmin`, `rmax` and `rstep` are understood as Å.
If the output file name starts with the string "kuplot", the output is not written to disk but placed into KUPLOT as last dat set.
No further output is written, unless the user explicitely states the optional parameters `iq:`, `sq:` and / or `fq:` with an appropriate output file name. These optional parameter cann all be stated on the same `output` command line or on individual lines as in the example.
Finally the operation is performed with the `run` command on line 14. As optional parameter `mode` you can specify "inter" or "silent", the latter being the default if the `mode:` parameter is omitted. With `mode:silent` DISCUS produces no further output until the menu is finished.
In interactive mode, intermediate data are displayed. In interactive mode, if the optional parameter `fourier:` has been omitted, DISCUS allows you to choose this value during the calculation.
Fig 2.1 shows the experimental intensity and background that is displayed in interactive mode. Note that the background has not yet been scaled. Fig 2.2 shows the final F(Q). The vertical red line mark the lower and upper limits. The upper limit $Q_{max;fourier}$ has already been adjusted to a position at which F($Q_{max;fourier}$) is zero.
The vertical green line marks the peak position used for the adjustment of the Q-scale. As DISCUS will search for the highest peak with a wide window of 0.25Å^{-1} width, choose the

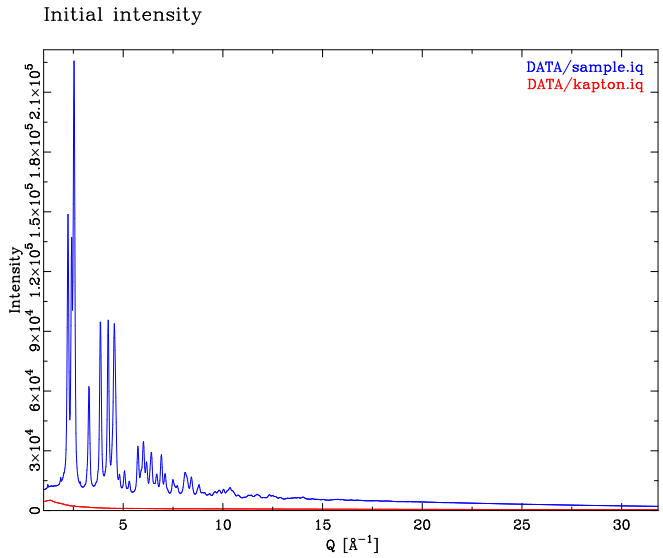

Figure 2.1: Initial intensity and background

highest maximum in the lower Q-range.
Finally Fig 2.3 shows the final G(r).

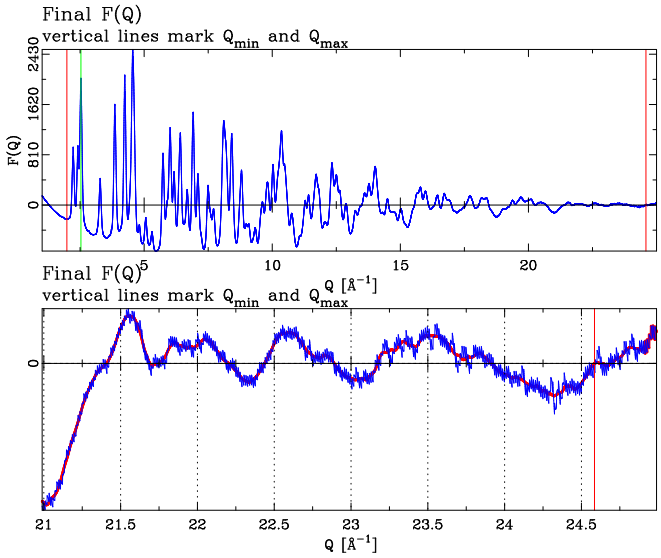

Figure 2.2: Final F(Q). The lower frame shows the last 4Å^{-1} to choose $Q_{max;fourier}$, The vertical green line shows the peak postion used for the optional adjustment of the Q-scale.

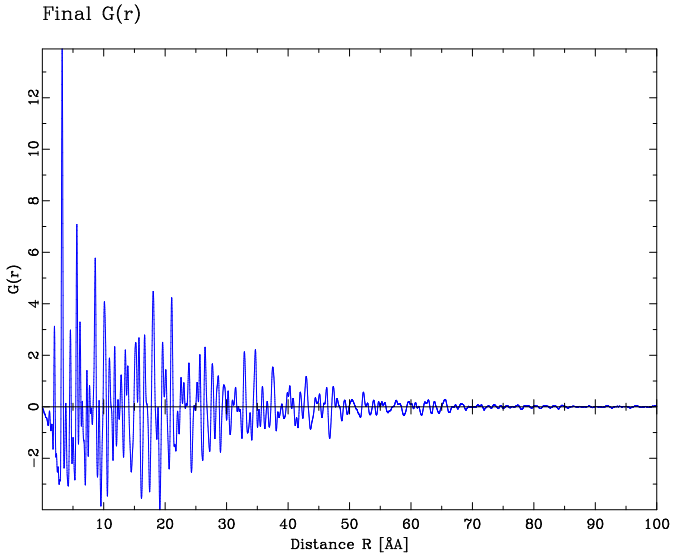

Figure 2.3: Final G(r). Note the absence of any Fourier ripples and the absence of any spurious peak in the low distance range.

Bibliography

S.J.L. Billinge; Ch. L. Farrow. J.Phys:Condens. Matter **25** (2013) 454202.