User manual of computer programs "RITS" for data analysis of Bragg-edge neutron transmission spectrum

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Introduction

RITS (Rietveld Imaging of Transmission Spectra) is computer program series which can quantitatively evaluate various crystalline microstructural information in the pulsed neutron Bragg-edge transmission imaging method. Owing to this data analysis code, the pulsed neutron Bragg-edge transmission imaging can become a useful material characterization tool. The aim of this user manual is progress of materials science using the pulsed neutron imaging by providing the information on how to use the RITS code, note and troubleshooting for users. The principle and the algorithm of the RITS code were discussed in:

H. Sato: "Quantitative Imaging of Crystalline Structure Information by using a Pulsed Neutron Transmission Method" Ph.D. Thesis, Hokkaido University, 2011. (In Japanese)

or

H. Sato, T. Kamiyama and Y. Kiyanagi: "A Rietveld-Type Analysis Code for Pulsed Neutron Bragg-Edge Transmission Imaging and Quantitative Evaluation of Texture and Microstructure of a Welded α-Iron Plate" Materials Transactions 52(6) (2011) 1294-1302. (In English)

Additionally, I also prepared a PPT file (**RITS_overview.ppt**) explaining the algorithm. Note that this user manual does not have the explanation of the algorithm.

Note

- **Program distribution by a user is forbidden without the permission by me. Please discuss with me.**
- Please **refer the five papers (see the next references)** when you present results obtained by the RITS code.

  [1] **H. Sato, T. Kamiyama and Y. Kiyanagi: "A Rietveld-Type Analysis Code for Pulsed Neutron Bragg-Edge Transmission Imaging and Quantitative Evaluation of Texture and Microstructure of a Welded α-Iron Plate" Materials Transactions 52(6) (2011) 1294-1302.** (Texture and Crystallite size)

  [2] **H. Sato, T. Shinohara, R. Kiyanagi, K. Aizawa, M. Ooi, M. Harada, K. Oikawa, F. Maekawa, K. Iwase, T. Kamiyama and Y. Kiyanagi: "Upgrade of Bragg edge analysis techniques of the RITS code for crystalline structural information imaging" Physics Procedia 43 (2013) 186-195.** (Crystal structure)

  [3] **H. Sato, T. Sato, Y. Shiota, T. Kamiyama, A. S. Tremsin, M. Ohnuma and Y. Kiyanagi: "Relation between Vickers Hardness and Bragg-Edge Broadening in Quenched Steel Rods Observed by Pulsed Neutron Transmission Imaging" Materials Transactions 56(8) (2015) 1147-1152.** (Bragg-edge shift (Macrostrain) and Bragg-edge broadening)

  [4] **H. Sato, K. Watanabe, K. Kiyokawa, R. Kiyanagi, K. Y. Hara, T. Kamiyama, M. Furusaka, T. Shinohara and Y. Kiyanagi: "Further Improvement of the RITS Code for Pulsed Neutron Bragg-edge Transmission Imaging" Physics Procedia 88 (2017) 322-330.** (Corrected crystal structure factor and crystallite size)

  [5] **H. Sato, M. Sato, Y. H. Su, T. Shinohara and T. Kamiyama: "Improvement of Bragg-edge neutron transmission imaging for evaluating the crystalline phase volume fraction in steel composed of ferrite and austenite" ISIJ International 61(5) (2021) 1584-1593.** (Crystalline phase volume fraction)

- Originally, the RITS code was developed for research and development of data analysis method of Bragg-edge neutron transmission data. In other words, this code is not user-friendly, and does not have enough data analysis speed. The GUI type high-speed RITS code is now developed by J-PARC MLF BL22 "RADEN" (the responsible person for software: Dr. Kenichi Oikawa).

- The RITS code adopted the non-linear least-squares analysis. Therefore, all the users, please confirm not only just the output data but also **the crystallographic/metallographic validity of the output data. The try& error iterations** are important. It is also important to **check the fitting curve by your eyes**.

- I would like to advice about your data analysis. Therefore, please send the neutron TOF transmission data and your "initial.dat" to me when you have a trouble.

<u>Special thanks for technical cooperation and testing</u>

Contents

1. Single Bragg-edge analysis program

The single Bragg-edge analysis program is a program for extraction of information on crystal lattice plane spacing and strain from single Bragg-edge. The 3 stage fitting method was adopted as well as ISIS (Santisteban *et al.*), but the edge profile function was changed to the Jorgensen-type function in the RITS code. The program consists of two types; edge1.f and edge2.f.

- edge1.f: analysis of single neutron TOF transmission data
- edge2.f: analysis of multiple position-dependent neutron TOF transmission data obtained by the imaging experiment

Hereafter, how to use these programs is presented in detail.

1.1 edge1.f

Please prepare:

- edge1.f   ⋯   source program (or its execute program "edge1.o")
- initial.dat   ⋯   initial crystalline microstructural parameter file before the fitting
- input.dat   ⋯   neutron TOF transmission data

Hereafter, detail of these files is presented.

1.1.1 input.dat

Fig. 1.1 shows a UNIX format of "input.dat" containing the neutron TOF-dependent transmission data.

   1st column: TOF (μs) (e.g.: bin width: 0 μs ~ 5 μs $\rightarrow$ 2.5 μs, bin width: 5 μs ~ 10 μs $\rightarrow$ 7.5 μs)
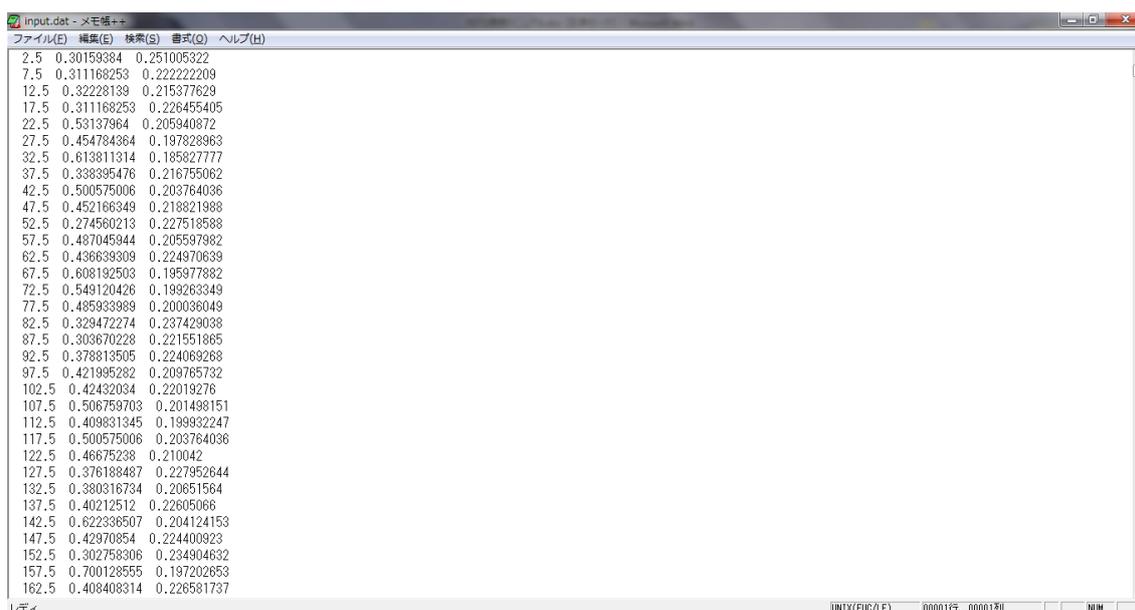
   2nd column: neutron transmission (e.g.: 0% $\rightarrow$ 0.0, 100% $\rightarrow$ 1.0)

   3rd column: statistics error (the standard deviation σ) of neutron transmission (0% $\rightarrow$ 0.0, 100% $\rightarrow$ 1.0)

"input.dat" can accept up to 16000 rows, which is limited by the RITS source program. However, 16000 is enough value because the 5 μs TOF analysis up to 8 Å can be done at the 40 m neutron flight path length beam-line under the condition of accelerator repetition of 25 Hz.

Various TOF value, linear/log setting and existence of skipped TOF bin are acceptable.

Recommended TOF bin width for strain analysis: 5 or 10 μs

Fig. 1.1: input.dat.

1.1.2 initial.dat

Fig. 1.2 shows a UNIX format of "initial.dat".

        1st row: a_0 in 23 page of "RITS overview.ppt" (0.0 is recommended)

        2nd row: b_0 in 23 page of "RITS overview.ppt" (0.0 is recommended)

        3rd row: a_{hkl} in 24 page of "RITS overview.ppt" (0.0 is recommended)

        4th row: b_{hkl} in 24 page of "RITS overview.ppt" (0.0 is recommended)

        5th row: d_{hkl} (Å) in 25 page of "RITS overview.ppt" (not $2d_{hkl}$)

        6th row: σ_{hkl} in 26 page or $\sigma_{0,hkl}$ in 30 page of "RITS overview.ppt" (2.0 ~ 10.0 are recommended)

        7th row: $\sigma_1'_{hkl}$ in 30 page of "RITS overview.ppt" (2.0 ~ 10.0 are recommended)

        8th row: α_{hkl} in 26 page of "RITS overview.ppt" (2.0 ~ 10.0 are recommended)

        9th row: β_{hkl} in 26 page of "RITS overview.ppt" (2.0 ~ 10.0 are recommended)

        10th row: starting point (rate for $2d_{hkl}$) of the 2nd stage fitting (shorter wavelength region)

        11th row: end point (rate for $2d_{hkl}$) of the 2nd stage fitting (shorter wavelength region)

        12th row: starting point (rate for $2d_{hkl}$) of the 3rd stage fitting (near-edge region)

        13th row: end point (rate for $2d_{hkl}$) of the 3rd stage fitting (near-edge region)

        14th row: starting point (rate for $2d_{hkl}$) of the 1st stage fitting (longer wavelength region)

        15th row: end point (rate for $2d_{hkl}$) of the 1st stage fitting (longer wavelength region)

        16th row: neutron flight path length from moderator to detector (m)

        17th row: facility setting parameter changing the order of σ_{hkl}, α_{hkl} and β_{hkl} (HUNS: 1, J-PARC MLF BL10 and BL22: 2, J-PARC MLF BL19: 3)

        18th row: 0: $\sigma_{0,hkl}$, $\sigma_1'_{hkl}$, α_{hkl} and β_{hkl} are constant; 1: $\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are variable ($\sigma_1'_{hkl}$ is constant); 2: $\sigma_1'_{hkl}$ is variable ($\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are constant).

        19th row: iteration trial number (3 or 2 is recommended)

Incidentally, comment at the end of each row indicates the unit and which the parameter is constant or refined or refinable.

Comment for 5th row

    d_{hkl} should be set a value near the value which you would like to investigate the crystal lattice plane spacing {hkl}. Note that is not the Bragg-edge wavelength ($\lambda_{hkl} = 2d_{hkl}$) but is the crystal lattice plane spacing (d_{hkl}). The RITS code automatically scans a Bragg-edge which should analyze based on this initial value.

Comment for 6th ~ 9th rows

    Data set of 1.0 should be avoided. The order of these parameters is changed by setting "facility setting parameter (17th row)".

Comment for 10th ~ 15th rows

    Fig. 1.3 shows scheme of these parameters. For example, if 14th row and 15th row are set;

        1.005   #14#   Start_position_1stFit[]constant

        1.01   #15#   End_position_1stFit[]constant

these represent a wavelength region of the 1st stage fitting of the 3 stage fitting. In this case, it means that the wavelength region λ

$$\lambda_{hkl} (= 2d_{hkl}) \times 1.005 < \lambda < \lambda_{hkl} (= 2d_{hkl}) \times 1.01$$

will be analyzed. As the same way,

        10th row & 11th row: set of wavelength region of the 2nd stage fitting

        12th row & 13th row: set of wavelength region of the 3rd (final) stage fitting

Incidentally, suitable values which I found are as follows:

Hokkaido University Neutron Source (HUNS): 0.9~0.95 / 0.95~1.2 / 1.2~1.4
J-PARC MLF BL10 (NOBORU) and BL22 (RADEN): 0.98~0.99 / 0.99~1.02 / 1.02~1.04
J-PARC MLF BL19 (TAKUMI): 0.994~0.999 / 0.9975~1.005 / 1.005~1.01

Here, note that before/after "~" means starting/end position value, and "/" separates 2nd / final / 1st stage, respectively.

Comment for 17th row

This parameter decides the order of $\sigma_{0,hkl}$, $\sigma_1'_{hkl}$, α_{hkl} and β_{hkl} of initial.dat (#6# ~ #9#).

Facility setting parameter "1" (HUNS): ×10.0 μs / ×0.01 μs^{-1} / ×0.001 μs^{-1}
Facility setting parameter "2" (NOBORU/RADEN): ×10.0 μs / ×0.01 μs^{-1} / ×0.01 μs^{-1}
Facility setting parameter "3" (TAKUMI): ×1.0 μs / ×0.01 μs^{-1} / ×0.01 μs^{-1}

These mean the order of σ / α / β, respectively. If σ (width) is smaller, or α (rise) and β (decay) are larger, Bragg-"edge" becomes sharp. The unit of σ, 1/α, 1/β is μs (microsecond).

Comment for 18th row and **Point for macrostrain/microstrain imaging**

This parameter can change refining/constant setting of $\sigma_{0,hkl}$, $\sigma_1'_{hkl}$, α_{hkl} and β_{hkl} of initial.dat (#6# ~ #9#).

0: $\sigma_{0,hkl}$, $\sigma_1'_{hkl}$, α_{hkl} and β_{hkl} are constant.

1: $\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are variable ($\sigma_1'_{hkl}$ is constant).

2: $\sigma_1'_{hkl}$ is variable ($\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are constant).

(1) **If you do not know your instrument resolution:** Firstly, you set the broadening setting parameter is "1", and only $\sigma_1'_{hkl}$ is zero. Then, analyze data of no Bragg-edge broadening (e.g. a μm-order grained material), and determine $\sigma_{0,hkl}$, α_{hkl} and β_{hkl} (instrument resolution).

(2) **If you know your instrument resolution, and hope macrostrain imaging without Bragg-edge broadening correction:** You set the broadening setting parameter is "0", $\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are set in suitable values (instrument resolution), and $\sigma_1'_{hkl}$ is zero. As a result, you can get refined d_{hkl} information without Bragg-edge broadening correction. This method is the most stable analysis decreasing the analysis error of d_{hkl}.

(3) **If you know your instrument resolution, and hope macrostrain and microstrain imaging:** You set the broadening setting parameter is "2", $\sigma_{0,hkl}$, α_{hkl} and β_{hkl} are set in suitable values (instrument resolution), and $\sigma_1'_{hkl}$ is set in 2.0~10.0. As a result, you can get refined d_{hkl} (relating to macrostrain) and $\sigma_1'_{hkl}$ (relating to microstrain) information with Bragg-edge broadening correction.
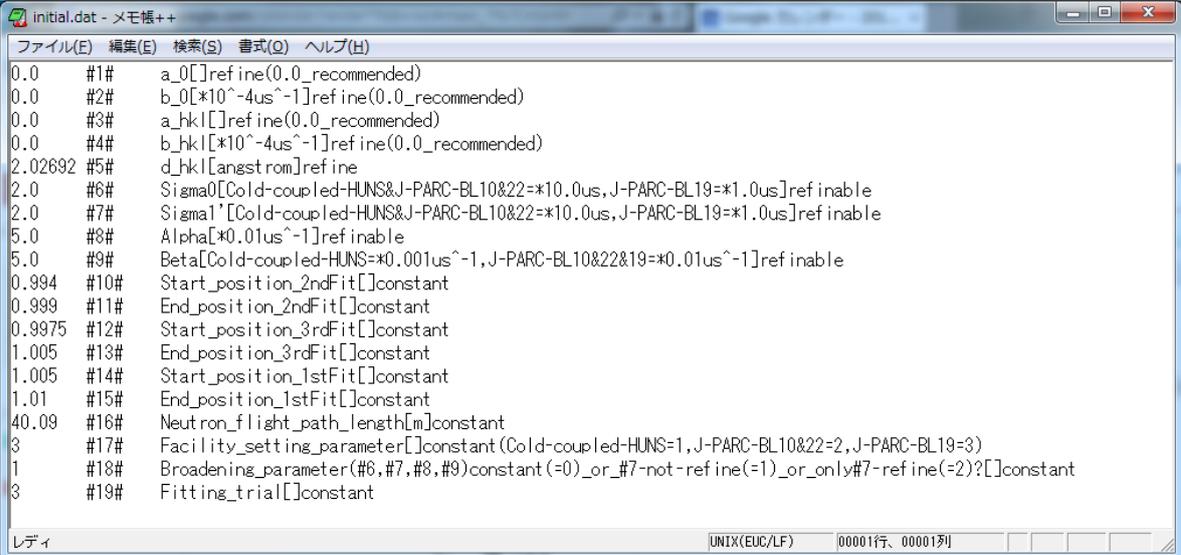
```
initial.dat - メモ帳++
ファイル(F)  編集(E)  検索(S)  書式(O)  ヘルプ(H)
0.0      #1#   a_0[]refine(0.0_recommended)
0.0      #2#   b_0[*10^-4us^-1]refine(0.0_recommended)
0.0      #3#   a_hkl[]refine(0.0_recommended)
0.0      #4#   b_hkl[*10^-4us^-1]refine(0.0_recommended)
2.02692  #5#   d_hkl[angstrom]refine
2.0      #6#   Sigma0[Cold-coupled-HUNS&J-PARC-BL10&22=*10.0us,J-PARC-BL19=*1.0us]refinable
2.0      #7#   Sigma1'[Cold-coupled-HUNS&J-PARC-BL10&22=*10.0us,J-PARC-BL19=*1.0us]refinable
5.0      #8#   Alpha[*0.01us^-1]refinable
5.0      #9#   Beta[Cold-coupled-HUNS=*0.001us^-1,J-PARC-BL10&22&19=*0.01us^-1]refinable
0.994    #10#  Start_position_2ndFit[]constant
0.999    #11#  End_position_2ndFit[]constant
0.9975   #12#  Start_position_3rdFit[]constant
1.005    #13#  End_position_3rdFit[]constant
1.005    #14#  Start_position_1stFit[]constant
1.01     #15#  End_position_1stFit[]constant
40.09    #16#  Neutron_flight_path_length[m]constant
3        #17#  Facility_setting_parameter[]constant(Cold-coupled-HUNS=1,J-PARC-BL10&22=2,J-PARC-BL19=3)
1        #18#  Broadening_parameter(#6,#7,#8,#9)constant(=0)_or_#7-not-refine(=1)_or_only#7-refine(=2)?[]constant
3        #19#  Fitting_trial[]constant
レディ                                                    UNIX(EUC/LF)   00001行、00001列
```
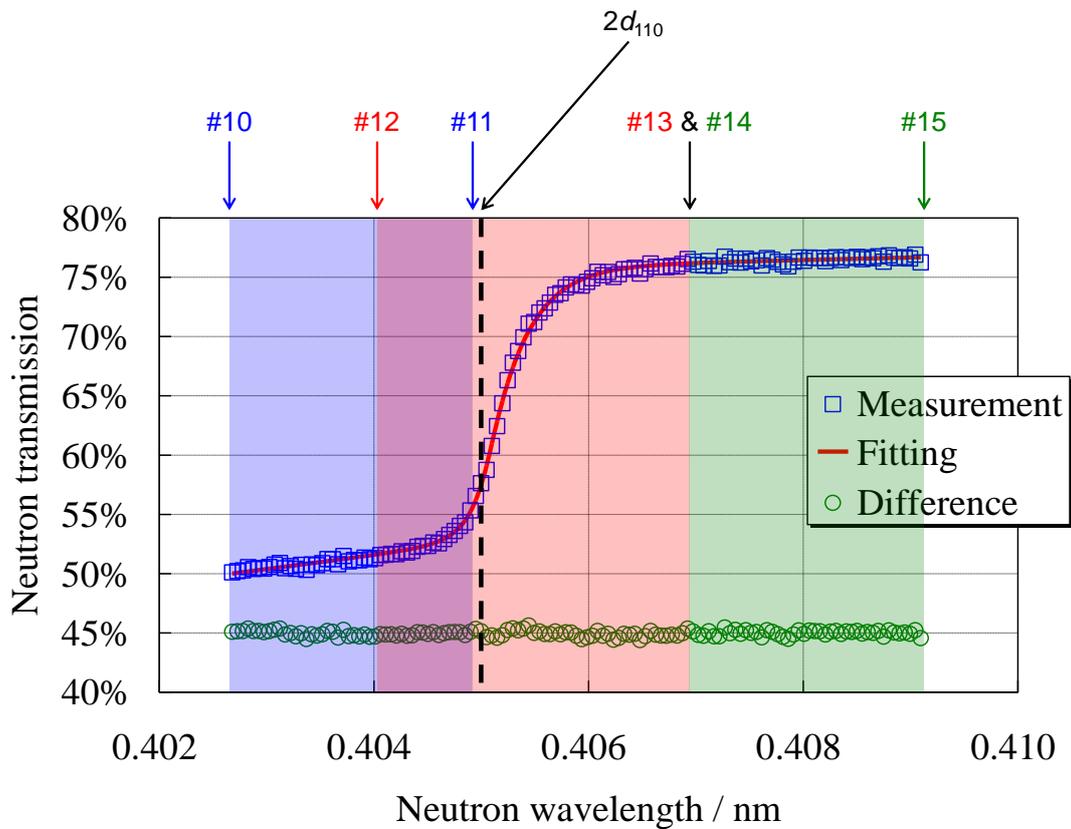
Fig. 1.2: initial.dat.

Fig. 1.3: Definition of #10#～#15# (analyzing region of 3 stage fitting).

1.1.3 edge1.f

Here, how to use the source program "edge1.f" is described. Firstly, input.dat, initial.dat and edge1.f are put at the same directly (folder). Then, the compile and the execution are done. Fig. 1.4 shows these commands. The compile command is

f77 edge1.f -o edge1.o

(if you already have the execution file "edge1.o", the above compile procedure can be skipped), and the execution command is

./edge1.o

After several seconds, ChiSq (χ^2 value) is displayed. That is all.

Fig. 1.4: Commands for check of files, the compile and the execution, and displayed χ^2 value.

Next, output data are explained. The output files are

- fit_para.dat　　　　　　$\cdots$　Refined crystalline microstructural parameters
- fit_func.dat　　　　　　$\cdots$　Fitting curve with experimental data

Fig. 1.5 shows "fit_para.dat". **Please read the sentences from the most bottom row of this file.** 1st column represents refined parameter, and 2nd column indicates its error (standard deviation). Each row represents as follows:

- Case 1: The broadening setting parameter (#18#): 0

    Final row: "Refined" parameter of 5th row in initial.dat (d_{hkl})

    1st row above final row: "Refined" parameter of 4th row in initial.dat (b_{hkl})

    2nd row above final row: "Refined" parameter of 3rd row in initial.dat (a_{hkl})

    3rd row above final row: "Refined" parameter of 2nd row in initial.dat (b_0)

    4th row above final row: "Refined" parameter of 1st row in initial.dat (a_0)

- Case 2: The broadening setting parameter (#18#): 1

    Final row: "Refined" parameter of 9th row in initial.dat (β_{hkl})

    1st row above final row: "Refined" parameter of 8th row in initial.dat (α_{hkl})

    2nd row above final row: "Refined" parameter of 6th row in initial.dat ($\sigma_{0,hkl}$)

    3rd row above final row: "Refined" parameter of 5th row in initial.dat (d_{hkl})

    4th row above final row: "Refined" parameter of 4th row in initial.dat (b_{hkl})

    5th row above final row: "Refined" parameter of 3rd row in initial.dat (a_{hkl})

    6th row above final row: "Refined" parameter of 2nd row in initial.dat (b_0)

    7th row above final row: "Refined" parameter of 1st row in initial.dat (a_0)

8

- Case 3: The broadening setting parameter (#18#): 2

    Final row: "Refined" parameter of 7th row in initial.dat ($\sigma_1{}'_{hkl}$)

    1st row above final row: "Refined" parameter of 5th row in initial.dat (d_{hkl})

    2nd row above final row: "Refined" parameter of 4th row in initial.dat (b_{hkl})

    3rd row above final row: "Refined" parameter of 3rd row in initial.dat (a_{hkl})

    4th row above final row: "Refined" parameter of 2nd row in initial.dat (b_0)

    5th row above final row: "Refined" parameter of 1st row in initial.dat (a_0)
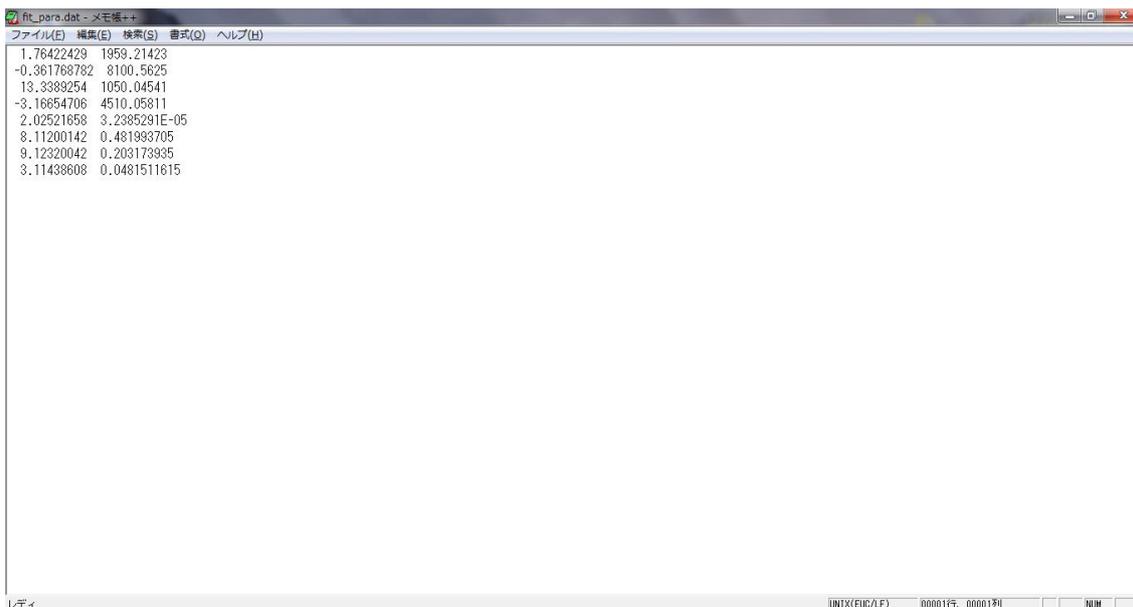
Fig. 1.5: fit_para.dat (case 2).

Fig. 1.6 shows "fit_func.dat".

    1st column: TOF (μs)

    2nd column: Wavelength from TOF (unit: Å, depending on #16# described in initial.dat)

    3rd column: Experimental data of neutron transmission (from input.dat)

    4th column: Fitting curve

    5th column: Difference between experimental data and fitting curve

Fig. 1.7 shows graph of "fit_func.dat". **Please confirm the validity of fitting analysis by your eyes.**

9

Fig. 1.6: fit_func.dat.

Fig. 1.7: Graph of fit_func.dat.

1.2　edge2.f

Please prepare:

- edge2.f   ⋯   source program
- initial.dat   ⋯   initial parameters file (the same one as edge1.f)
- data.dat   ⋯   including all input files name like "input.dat", which you want to analyze
- Input files like "input.dat"

1.2.1　data.dat

Fig. 1.8 shows contents of "data.dat". Many files name are described in this file; 1-1.dat, 1-2.dat, 1-3.dat, ⋯. These

files (1-1.dat, 1-2.dat, 1-3.dat, ⋯) are the same ones as "input.dat" for edge1.f. In other words, many "input.dat"-like files can be successively analyzed, one after another, by read this file (data.dat). Therefore, of course, files described in "data.dat" (1-1.dat, 1-2.dat, 1-3.dat, ⋯) must be prepared. So-called full-path setting is capable, therefore readout from a USB memory is also possible. 65536 (256×256) of input files are acceptable at once.
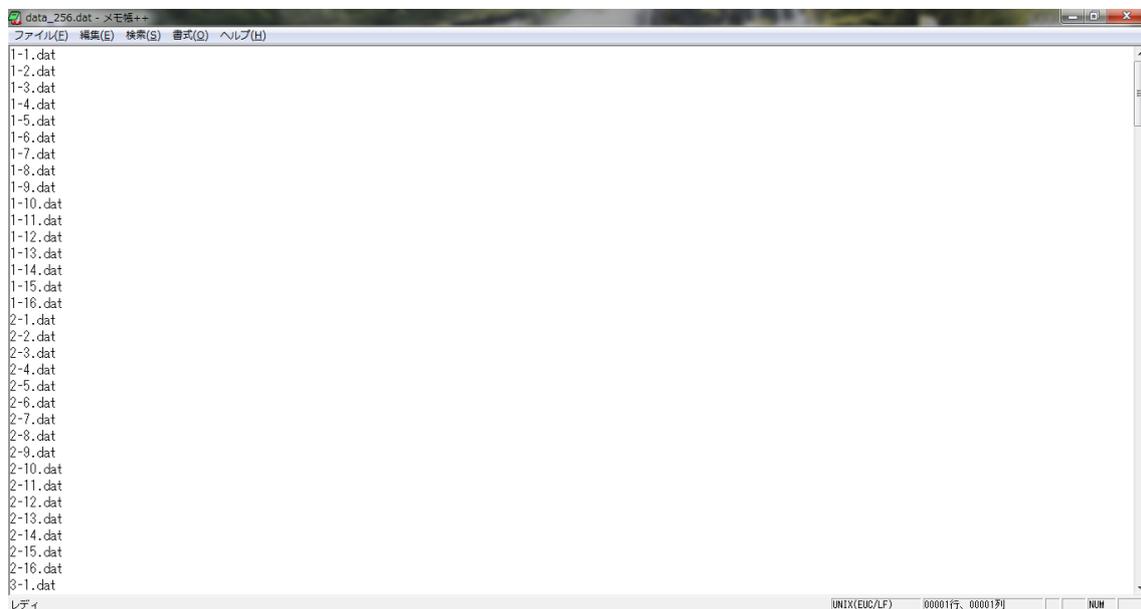
Fig. 1.8: data.dat.

1.2.2 initial.dat

This is the same file explained in Sec. 1.1.2. It is better to use the refined typical parameters obtained by edge1.f, as initial parameters for edge2.f.

1.2.3 edge2.f

The compile and the execution should be done. Hereafter, I introduce a convenient technique for edge2.f. Fig. 1.9 shows a result after the "normal" execution (see Sec. 1.1.3). The refined parameters are displayed in the monitor. However, in this case, we cannot preserve the refined parameters as a text file. On the other hand, please command

./edge2.o **> result.dat**

where "result.dat" (your arbitrary name) is the file name preserving the output data which would be displayed in the monitor. Thus, we can preserve the output data as a text data (see Fig. 1.10).

The output result consists of as follows.

- Case 1: The broadening setting parameter (#18#): 0

  1st column: refined d_{hkl}

  2nd column: error/standard-deviation of d_{hkl}

  3rd column: χ^2 value

- Case 2: The broadening setting parameter (#18#): 1

  1st column: refined d_{hkl}

  2nd column: error/standard-deviation of d_{hkl}

  3rd column: refined $\sigma_{0,hkl}$

  4th column: refined α_{hkl}

11

5th column: refined β_{hkl}

6th column: χ^2 value

- Case 3: The broadening setting parameter (#18#): 2

    1st column: refined d_{hkl}

    2nd column: error/standard-deviation of d_{hkl}

    3rd column: refined σ_1'$_{hkl}$

    4th column: error/standard-deviation of σ_1'$_{hkl}$

    5th column: χ^2 value

Incidentally, obtained results are the same ones as edge1.f.

Fig. 1.9: Normal execution of edge2.o.

Fig. 1.10: Execution of edge2.o which we set a certain output file name.

2. Rietveld-type Bragg-edge analysis program

The Rietveld-type Bragg-edge analysis program was developed for quantitative analysis of texture and crystallite size from whole pattern of Bragg-edge transmission spectrum. The program series consist of rits1.f and rits2.f.

- rits1.f: analysis of single neutron TOF transmission data
- rits2.f: analysis of multiple position-dependent neutron TOF transmission data obtained by the imaging experiment

Hereafter, how to use these programs is presented in detail.

2.1 rits1.f

rits1.f has two functions. One is fitting analysis of experimental data, and the other is simulation calculation of total cross-section data. Please prepare:

- rits1.f   ⋯   source program
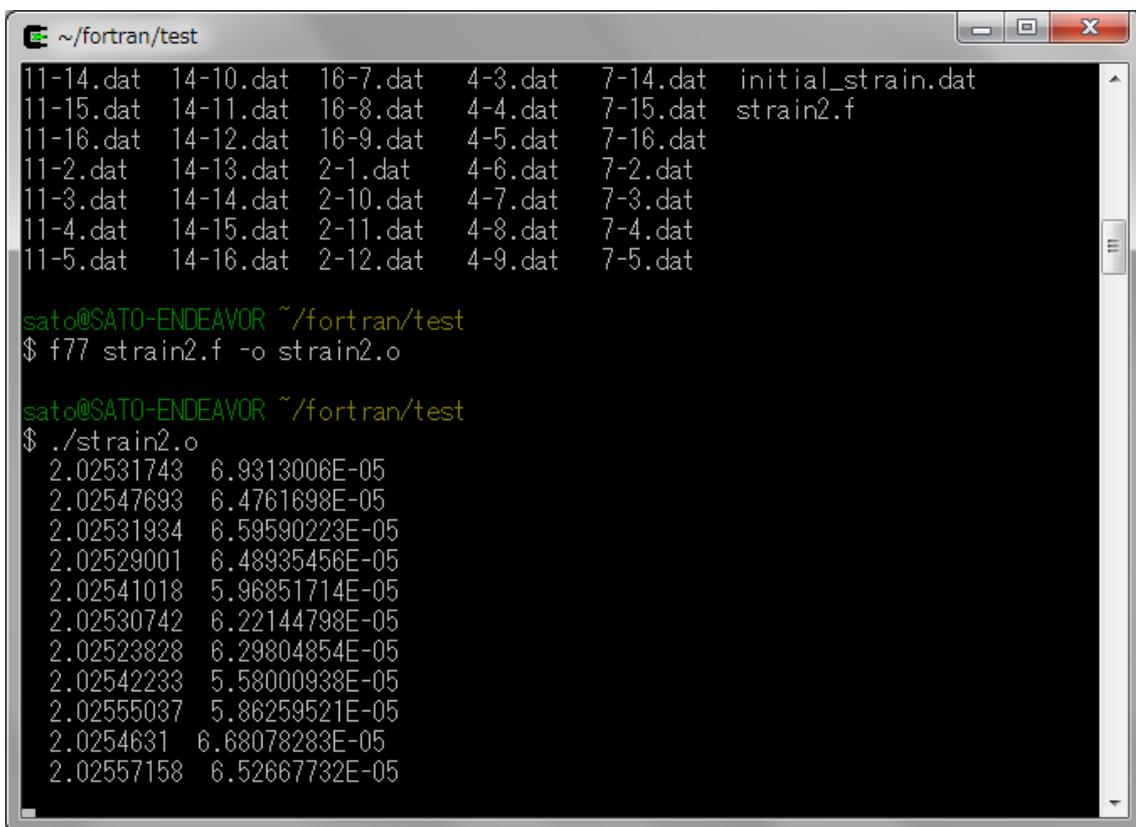- initial.dat   ⋯   initial data file of crystalline microstructure
- spgra   ⋯   Space group database
- input.dat   ⋯   Neutron TOF-dependent transmission spectrum data (in case of simulation of total cross-section, this file is not needed.)

2.1.1 input.dat

This is the same file as one explained in Sec. 1.1.1 Incidentally, this file is not needed in case of simulation calculation mode of total cross-section.

Recommended TOF bin width for texture and crystallite size analysis (almost proportional to the neutron flight path length):

50 μs (at HUNS (6 m))

100 μs (at J-PARC MLF BL10 "NOBORU" (14 m))

200 μs (at J-PARC MLF BL19 "TAKUMI" (40 m))

This is because too much TOF bin number causes slow data analysis speed.

2.1.2 spgra

Please put this file at the same directory (folder).

2.1.3 initial.dat

Refinable parameter is arbitrary parameter indicated by described as "refinable" in initial.dat. If you refine a certain parameter, please add "? " mark in front of the value as follows.

? 2.8665   #13#   Crystal_lattice_constant_parameter_a[angstrom]refinable

Space after "?" is necessary. Parameters with "?" will be refined, but parameters without "?" will not be refined (constant).

【Whole structure of "initial.dat"】

Parameters for experimental/analysis condition

   Parameters for crystalline phase No. 1

     Parameters for characteristics of phase No. 1

       Parameters for atom No. 1 in phase No. 1

       Parameters for atom No. 2 in phase No. 1

       …

<div align="center">

Parameters for crystalline phase No. 2

Parameters for characteristics of phase No. 2

Parameters for atom No. 1 in phase No. 2

Parameters for atom No. 2 in phase No. 2

…

</div>

These parameters are identified by the number of row from the most upper side in initial.dat. Therefore, you can delete **domains** about "**phase**" and "**atom**" which you do not use in inititial.dat. **But, you must not delete rows which are needed for analysis (e.g., first 7 rows, and information on space group, density, lattice parameters, edge profile, March-Dollase function, extinction function, the number of atoms of the phase).**

【1st row】

Facility setting parameters. These are the same parameters as ones of edge.f series. The edge profile function is affected by these parameters.

【4th and 5th rows】

4th row: the first number of TOF channel which you would like to analyze.

5th row: the last number of TOF channel which you would like to analyze.

Note that unit of them is not μs or Å, but is channel number.

【6th row】

The same effect as the edge.f series. 2 or 3 are recommended.

【7th row】

The number of crystalline phase, up to "2". If you set "1", you can delete all parameters for 2nd crystalline phase.

【8th ~ 11th rows】

Parameters for readout from the space group data file "spgra". As a result, the primitive unit lattice can be formed in the RITS code.

8th row: Normally, 0.

9th row: For example, 229 (BCC), 225 (FCC) and 194 (HCP).

10th row: Normally, 1.

11th row: Coordination number; e.g., 2.0 (BCC and HCP) or 4.0 (FCC)

About the detail, please refer and the crystallographic data base. I recommend:

http://www.crystallography.net/

【12th row】

Projection data of atomic number density (atomic number density × thickness or so-called atomic number area density). **Refine is recommended.** Incidentally, the ideal value of **alpha-iron of 5 mm thickness** is **4.24553** ($\times 10^{22}$ cm^{-2}), but the actual value is smaller than the ideal value.

【13th ~ 18th rows】

Crystal lattice constant parameters. The fitting analysis under the below settings is worked. Note that this function about length parameters (#13#, #14# and #15#) case is also applicable to angle parameters (#16#, #17# and #18#) case.

- Case 1: If you analyze a crystal structure of #13# = #14# = #15# (e.g., BCC or FCC):

Please set #13# = #14# = #15# and #13#, and only #13# is refined as follows:

? 2.8665	#13#	Crystal_lattice_constant_parameter_a[angstrom]refinable
2.8665	#14#	Crystal_lattice_constant_parameter_b[angstrom]refinable
2.8665	#15#	Crystal_lattice_constant_parameter_c[angstrom]refinable

As a result, #13# and #14# and #15# are simultaneously refined under the condition that these have the same value. Note that you should describe quite same value in this case.

- Case 2: If you analyze a crystal structure of #13# = #14#, but #15# is different from others (e.g., HCP):

Please set #13# = #14#, but #15# is different value from others, and #13# and #15# are refined as follows:

? 2.8665	#13#	Crystal_lattice_constant_parameter_a[angstrom]refinable
2.8665	#14#	Crystal_lattice_constant_parameter_b[angstrom]refinable
? 1.8665	#15#	Crystal_lattice_constant_parameter_c[angstrom]refinable

As a result, #13# and #14# are simultaneously refined under the condition that only #13# and #14# have the same value.

- Case 3: If you analyze a crystal structure of different values of #13# and #14# and #15#.

Please set different parameters in all, and refine all the parameters by three "?". Note that you should describe different values in this case.

【19th row】

"1" means the edge profile function is the Jorgensen-type resolution function. "0" means the edge profile function is the Heaviside's step function (the edge broadening does not exist), and parameters described in this edge-profile section do not work.

【20th row and 21st row】

Wavelength range of the edge profile function. These are the same parameters as edge.f, also correspond to the setting of #10# and #15# of Fig. 1.3, respectively.

【22nd ~ 28th rows】

If you set the 19th row is "1", please input suitable values here. I recommend that #22# and #25# and #27# are set in the same values as instrument resolution refined by edge1.f ($\sigma_{0,hkl}$ and α_{hkl} and β_{hkl}), and these parameters are constant during the RITS analysis. This is because refinement of these parameters is difficult for rits.f (affection from whole pattern (spectrum) is too strong), and accuracy and precision of determination of these parameters are higher in the single edge analysis and the Pawley-type analysis than the Rietveld-type analysis.

【29th row】

In this program, three March-Dollase functions can be combined. Please set the number of functions (0~3) here. However, "0" or "1" are recommended because the calculation speed is too slow in case of "2" and "3". In case 0 is set, there is not texture, and the calculation speed is faster than the case of using the March-Dollase function. For the texture analysis, we usually select "1" in this row.

【30th row and 31st row】

If you set "1" as the 29th row, the contribution of the first (single) March-Dollase function is automatically set as 1.0 (100%).

If you set "2" as the 29th row, you have to set a certain contribution of the first March-Dollase function in the 30th row (the contribution of the second March-Dollase function is automatically set as "1.0 - (the contribution of the first March-Dollase function)").

If you set "3" as the 29th row, you have to set the 30th row (the contribution of the first March-Dollase function) and the 31st row (the contribution of the second March-Dollase function). The contribution of the third March-Dollase function is automatically calculated in the program.

【32nd ~ 34th rows】

March-Dollase coefficients. The setting of "1.0" is forbidden when you refine the March-Dollase coefficient. About the effects, please see 37 page of "RITS overview.ppt".

【35th ~ 43rd rows】

H, K and L of preferred orientation vector <HKL> are integer. In the condition that ratio among H, K and L is same, the same March-Dollase function will be calculated because HKL is the Miller index of direction.

From 35th row to 37th row are preferred orientation vector of 1st March-Dollase function, from 38th row to 40th row are preferred orientation vector of 2nd March-Dollase function, and from 41st row to 43rd row are preferred orientation vector of 3rd March-Dollase function.

【44th row】

Only the coherent elastic scattering total cross-section having Bragg-edges appearing at longer wavelength region than this wavelength (Å) (44th row) will be multiplied by the March-Dollase function. Note that if too wide wavelength range was set, the calculation time becomes too long. Please set a reasonable wavelength region.

【45th row】

Crystallite size (μm). The recommended initial value for fitting is about 2.0.

【46th row】

Under the same principle as the 44th row, the wavelength region multiplied by the primary extinction effect function is set. However, about this function, if the wavelength region becomes wider, the calculation time does not become too long.

【47th row】

The number of element in the crystalline phase No. 1 is set.

【48th row】

Site occupancy. The principle is same as the Rietveld analysis codes such as GSAS, FullProf, RIETAN and Z-Rietveld.

【49th row ~ 51st row】

Fractional coordinates (x, y, z). The database for crystallography

        http://www.crystallography.net/

should be referred. Incidentally,

        BCC (No. 229): 0.0, 0.0, 0.0
        FCC (No. 225): 0.0, 0.0, 0.0
        HPC (No. 194): 0.3333333333, 0.6666666666, 0.25

【52nd row and 53rd row】

Isotropic atomic displacement parameters (see page 13 and page 14 of RITS overview.ppt). If you want to know reasonable values of them, rits1.f is executed under the simulation mode, and you can watch the theoretical values in your PC monitor (see Fig. 2.2). Please use these theoretical values as initial values for fitting.

If you set these rows are 0.0, these parameters cannot be refined. However, simulation calculation of total cross section and profile fitting analysis can be performed by using the theoretical (constant) values automatically calculated in RITS.

On the other hand, if you want to refine the isotropic atomic displacement parameters, the theoretical values outputted in the PC monitor above the method (Fig. 2.2) are recommended as the initial values of initial.dat.

【54th row ~ 59th row】

Please input nuclear data and atom data. Please refer the NIST database

https://www.ncnr.nist.gov/resources/n-lengths/

【Hereafter】

The above explanation is for 1st element of 1st crystalline phase. Please follow the structure of initial.dat as explained at the first of Sec. 3.1.3, and set another element/phase information.

【Point for better fitting】

You should avoid that the number of refinement parameters increases as possible as you can. **Projected atomic number density**, **March-Dollase coefficient** and **crystallite size** are recommended as refinement parameter. **Crystal lattice constant parameter** is also recommended. This is because these parameters are sensitive to vertical position, shape, intensity and horizontal position of the neutron transmission spectrum, respectively.

2.1.4 rits1.f

Fig. 2.1 shows a PC monitor after the compile/execution of rits1.f. Here, the RITS program ask you

Rietveld refinement ? (y / n)

Please reply "y" (Yes) or "n" (No) (input/enter). In case of "y", RITS carries out profile fitting analysis toward experimental data (input.dat). In case of "n", RITS carries out simulation calculation of total cross-section.

Fig. 2.2 shows a PC monitor after the execution of the simulation calculation mode of total cross-section. Isotropic atomic displacement parameter and $\varphi_1(\Theta) \times \varphi_3(\Theta)$ are displayed. These values can be used for initial values of 52nd row and 53rd row of initial.dat.

Fig. 2.1: rits1.o executed.

Fig. 2.2: Isotropic atomic displacement parameter B_{iso} ($\times 0.1$ Å^2) and $\varphi_1(\Theta) \times \varphi_3(\Theta)$ ($\times 0.1$) displayed after the RITS execution (simulation mode). This example is Fe of 293.6 K ($B_{\mathrm{iso}} = 0.29$ Å^2 and $\varphi_1(\Theta) \times \varphi_3(\Theta) = 0.63$).

If "n" is selected, "simulation.dat" will be outputted. Fig. 2.3 shows simulation.dat (single crystalline phase case). Fig. 2.4 shows a graph of simulation.dat (single crystalline phase case)

  1st column: Neutron wavelength (Å)

  2nd column: Coherent elastic scattering cross section (barn)

  3rd column: Incoherent elastic scattering cross section (barn)

  4th column: Coherent inelastic scattering cross section (barn)

  5th column: Incoherent inelastic scattering cross section (barn)

  6th column: Absorption cross section (barn)

Note that total cross section is not outputted. Please sum these cross section components by yourself.

  On the other hand, double crystalline phase case:

  1st column: Neutron wavelength (Å)

  2nd column: Total cross section of 1st crystalline phase (barn)

  3rd column: Total cross section of 2nd crystalline phase (barn)

  4th column: Sum of total cross sections of 1st and 2nd crystalline phases (barn)

simulation.dat - メモ帳++

ファイル(F) 編集(E) 検索(S) 書式(O) ヘルプ(H)

```
0.5000000E-02  0.4701049E-03  0.1709656E-04  0.1082493E+02  0.3859157E+00  0.7119021E-02
0.1500000E-01  0.4230836E-02  0.1538691E-03  0.1082111E+02  0.3857791E+00  0.2135706E-01
0.2500000E-01  0.1175251E-01  0.4274141E-03  0.1081345E+02  0.3855061E+00  0.3559510E-01
0.3500000E-01  0.2303484E-01  0.8377315E-03  0.1080196E+02  0.3850965E+00  0.4983314E-01
0.4499999E-01  0.3807708E-01  0.1384821E-02  0.1078664E+02  0.3845504E+00  0.6407118E-01
0.5499999E-01  0.5668171E-01  0.2068684E-02  0.1076749E+02  0.3838677E+00  0.7830922E-01
0.6499999E-01  0.7944439E-01  0.2889319E-02  0.1074451E+02  0.3830485E+00  0.9254726E-01
0.7499999E-01  0.1057713E+00  0.3846726E-02  0.1071770E+02  0.3820929E+00  0.1067853E+00
0.8499999E-01  0.1358574E+00  0.4940906E-02  0.1068707E+02  0.3810006E+00  0.1210233E+00
0.9499998E-01  0.1697036E+00  0.6171858E-02  0.1065260E+02  0.3797718E+00  0.1352614E+00
0.1050000E+00  0.2073134E+00  0.7539583E-02  0.1061430E+02  0.3784065E+00  0.1494994E+00
0.1150000E+00  0.2486798E+00  0.9044080E-02  0.1057218E+02  0.3769047E+00  0.1637375E+00
0.1250000E+00  0.2938155E+00  0.1068535E-01  0.1052622E+02  0.3752663E+00  0.1779755E+00
0.1350000E+00  0.3426983E+00  0.1246339E-01  0.1047643E+02  0.3734914E+00  0.1922135E+00
0.1450000E+00  0.3953418E+00  0.1437821E-01  0.1042282E+02  0.3715800E+00  0.2064516E+00
0.1550000E+00  0.4517650E+00  0.1642980E-01  0.1036537E+02  0.3695321E+00  0.2206896E+00
0.1650000E+00  0.5119312E+00  0.1861816E-01  0.1030410E+02  0.3673476E+00  0.2349277E+00
0.1750000E+00  0.5758710E+00  0.2094329E-01  0.1023900E+02  0.3650266E+00  0.2491657E+00
0.1850000E+00  0.6435559E+00  0.2340520E-01  0.1017006E+02  0.3625690E+00  0.2634038E+00
0.1950000E+00  0.7150147E+00  0.2600387E-01  0.1009730E+02  0.3599750E+00  0.2776418E+00
0.2050000E+00  0.7902343E+00  0.2873931E-01  0.1002071E+02  0.3572444E+00  0.2918799E+00
0.2150000E+00  0.8692235E+00  0.3161145E-01  0.9940285E+01  0.3543773E+00  0.3061179E+00
0.2250000E+00  0.9519537E+00  0.3462022E-01  0.9856037E+01  0.3513739E+00  0.3203560E+00
0.2350000E+00  0.1038427E+01  0.3776537E-01  0.9767973E+01  0.3482343E+00  0.3345940E+00
0.2450000E+00  0.1128715E+01  0.4104646E-01  0.9676102E+01  0.3449591E+00  0.3488321E+00
0.2550000E+00  0.1222734E+01  0.4446266E-01  0.9580446E+01  0.3415489E+00  0.3630701E+00
0.2650000E+00  0.1320488E+01  0.4801267E-01  0.9481047E+01  0.3380052E+00  0.3773081E+00
0.2750000E+00  0.1422019E+01  0.5169449E-01  0.9377954E+01  0.3343299E+00  0.3915462E+00
0.2850000E+00  0.1527284E+01  0.5550532E-01  0.9271252E+01  0.3305259E+00  0.4057842E+00
0.2950000E+00  0.1636154E+01  0.5944142E-01  0.9161040E+01  0.3265968E+00  0.4200222E+00
0.3050000E+00  0.1748619E+01  0.6349805E-01  0.9047457E+01  0.3225475E+00  0.4342602E+00
0.3150000E+00  0.1864618E+01  0.6766940E-01  0.8930660E+01  0.3183836E+00  0.4484983E+00
0.3250000E+00  0.1983712E+01  0.7194867E-01  0.8810844E+01  0.3141121E+00  0.4627363E+00
0.3349999E+00  0.2105943E+01  0.7632811E-01  0.8688226E+01  0.3097407E+00  0.4769743E+00
```
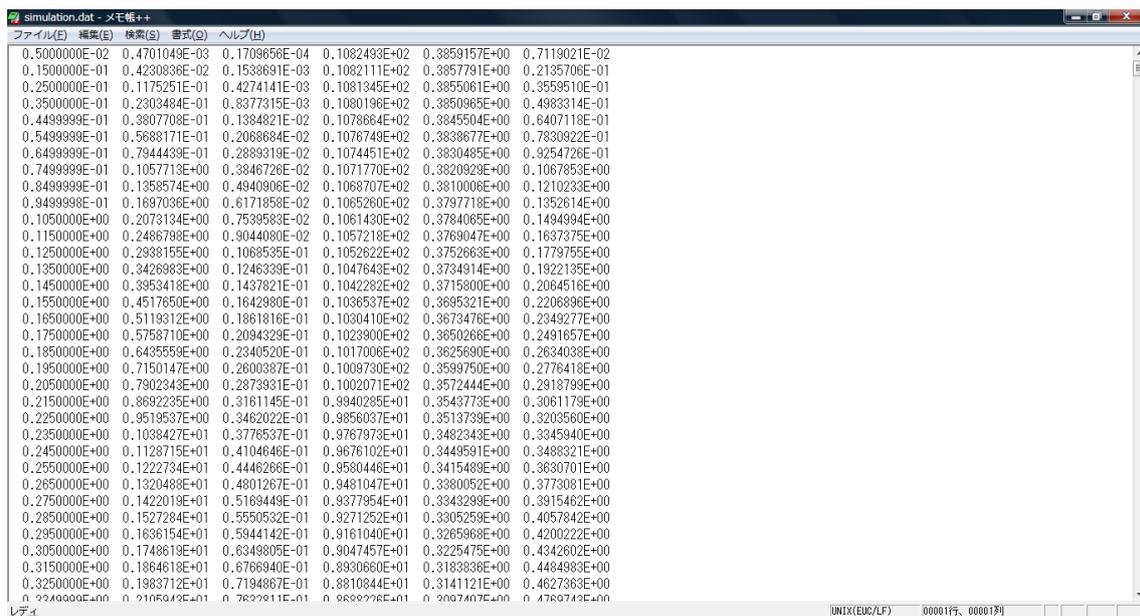
UNIX(EUC/LF)   00001行、00001列
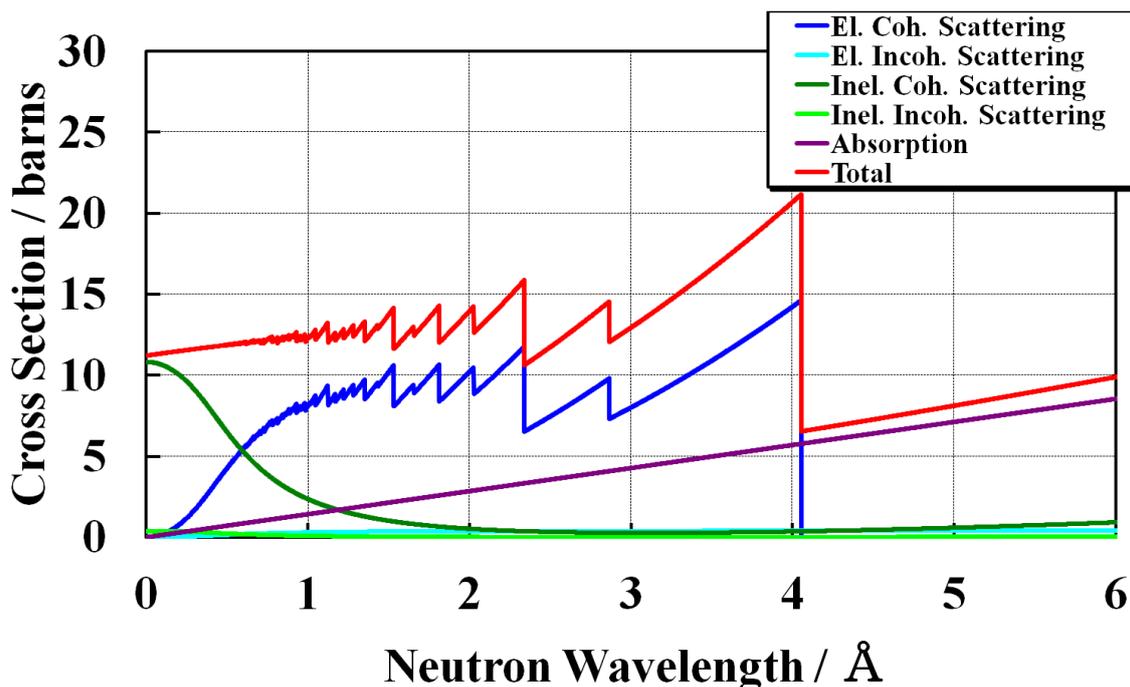
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Fig. 2.3: simulation.dat.

Fig. 2.4: Graph of simulation.dat.

If you select "y", the profile fitting analysis is run (see Fig. 2.5). The outputted values are current status of refined parameters indicated by "?" in initial.dat. If these parameters are out of control, please do forced termination by the command "ctrl + c", and improve your initial.dat. The output monitor shows sometimes

"trial number" "converging number" "χ^2 value" (2 2 80.4640427 in Fig. 2.5)

Then, finally, χ^2 value is also displayed.

In case the fitting analysis that the trial number is equal to the converging number is done, fitting is too good or too

bad. In case of the latter case, please improve your initial.dat. In this case, **the crystallographic/metallographic validity of the initial data** and **the calculated transmission spectrum** should be checked **by your knowledge and eyes**. It is also important to check a result of simulation calculation of total cross-section and its failure.

After the fitting analysis, fit_para.dat and fit_func.dat are outputted. fit_func.dat corresponds to that explained at Sec. 1.1.3. fit_para.dat almost corresponds to that explained at Sec. 1.1.3, however, **each row quite corresponds to the row set in initial.dat**.

Fig. 2.5: PC monitor after the execution of profile fitting analysis.

2.2 rits2.f

Please prepare:

- rits2.f　　　　　　⋯ source program
- initial.dat　　　　　⋯ initial parameters file
- spgra　　　　　　　⋯ space group database
- data.dat　　　　　　⋯ including all input files name like "input.dat", which you want to analyze
- Input files like "input.dat"

How to use "rits2.f" will be understood if you know how to use "edge2.f" and "rits1.f". Note that the outputted data are only **1st, 2nd, 3rd, 4th, ... 10th column refined parameters, and χ^2 value**. If you also need 11th, 12th, … data, please contact me. Incidentally, if you set 7 as the number of refinement parameter, 8th, 9th 10th column output data should be ignored.

data.dat can accept up to 4096 input.dat-like data files.