



# User guide to VESUVIO data analysis programs for powders and liquids

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# **User guide to VESUVIO data analysis**

## **programs for powders and liquids**

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## 1. Introduction

This guide considers the analysis of VESUVIO data from samples with no preferred orientation- for example powders, liquids, glasses, amorphous materials etc. By following the procedures given the user should be able to fully analyse the data from any such sample. The theory behind the analysis is outlined in more detail in Appendix 2. However the main features of the data analysis are as follows.

VESUVIO time of flight spectra consist of a series of peaks. Each peak corresponds to a specific atomic mass  $M$  in the sample studied. Positions of the peaks are uniquely determined by  $M$ . The amplitude of a peak is determined by the number of atoms of mass  $M$  and the scattering cross-section. The shape and width of the peak is determined by the momentum distribution  $n(p)$  of atoms of mass  $M$ . The ultimate aim of the data analysis is to determine the shape and intensity of the peaks corresponding to different masses and hence the momentum distributions of the atoms and the composition of the sample.

The programs are illustrated throughout by the analysis of a measurement on Zirconium Hydride ( $\text{ZrH}_{1.98}$ ). This consisted of seven runs (14188-14195) with a total of 6500  $\mu\text{amp}$ -hours.  $\text{ZrH}_2$  is a strongly scattering sample and the run time is considerably longer than that of a typical run ( $\sim 2000$   $\mu\text{amp}$ -hours) hence this data provides a stringent test of the analysis procedures.

The data for a single detector at a scattering angle of  $52^\circ$ , after the correction procedure described in section 3, is shown in Figure 1. There are three peaks in the data (1) Hydrogen gives a peak centred at  $\sim 245.6$   $\mu\text{sec}$ . (2) Zirconium gives a peak centred at  $382.3$   $\mu\text{sec}$ . (3) The aluminium can gives a peak centred at  $378.7$   $\mu\text{sec}$ . The latter two peaks are not resolved at forward scattering.

The corresponding data from a single detector in the back scattering bank at angle of  $163^\circ$  is shown in Figure 2. In this case there is no hydrogen peak. At eV neutron energies (to a good approximation see Appendices 2 and 3), the neutron scatters from single atoms and kinetic energy and momentum are conserved in the scattering process. Neutrons cannot back-scatter in a single scattering from a proton as they have almost the same mass, just as one billiard ball cannot back-scatter from another. The zirconium peak is centred at  $366.4$   $\mu\text{sec}$ , while the aluminium peak is centred at  $348.8$   $\mu\text{sec}$ . As the scattering angle increases, the peaks move further apart in the spectra. Hence at back-scattering the fact that there are separate peaks due to aluminium and zirconium is apparent in the time of flight data.

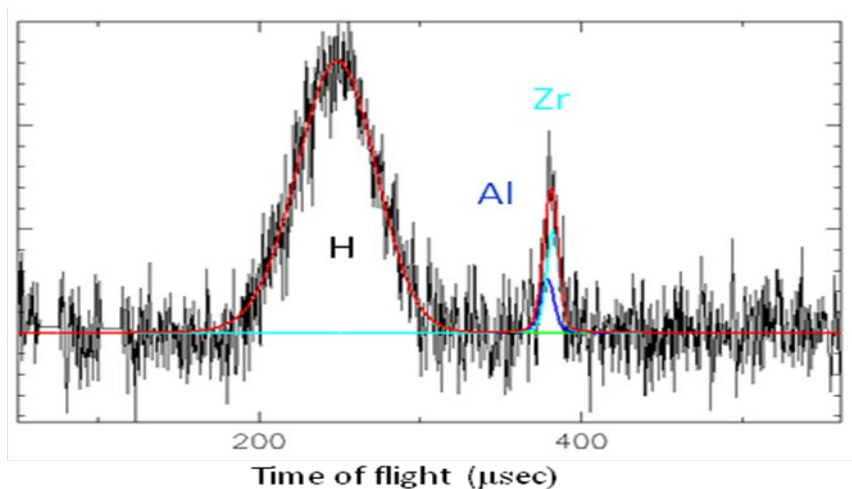


Figure 1. Time of flight data from Zirconium Hydride at a scattering angle of  $52^\circ$ . The data is shown after the corrections described in section 3. The fit obtained using the TFIT routine is shown as the red line. The individual peaks contributing to the fit are also shown. The hydrogen peak coincides with the red line and hence is not visible.

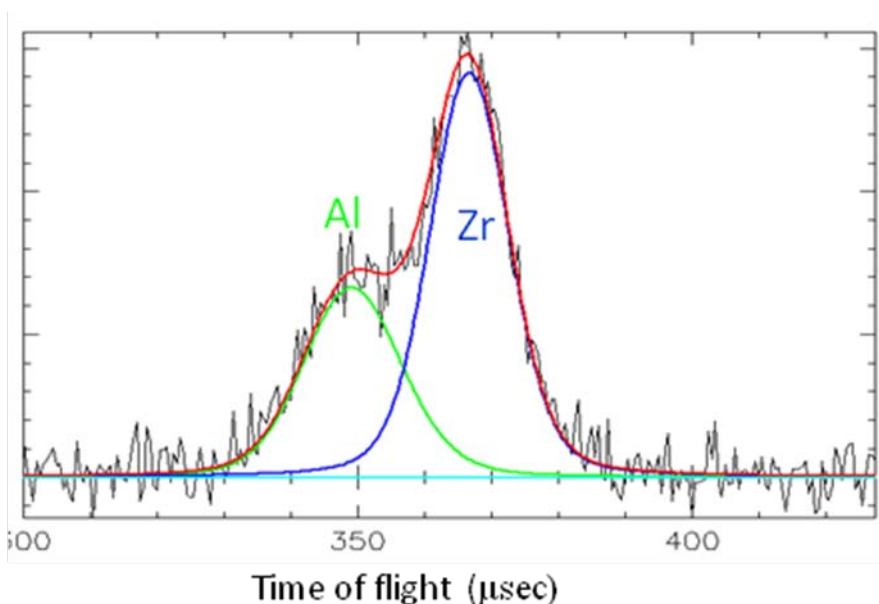


Figure 2. Time of flight data from Zirconium Hydride at a scattering angle of  $163^\circ$  degrees. The red line is a fit to the two peaks using the TFIT command. The green line is the contribution of the aluminium can. The blue line is the zirconium contribution.

Throughout the text we adopt the convention that **input from the user** is in red **Output from the programs** is in blue. Explanations are in black. None of the commands are case sensitive.

The commands in the manual can be set up in your own user account by the following command when you login for the first time

```
copy evs$disk0:[evsmgr.userprogs]login.com *
```

Then

```
@login
```

To set up your user account you need to fill in a form which can be printed by clicking on the “computer account form” pdf icon on the desktop of the instrument computer in the VESUVIO cabin. This has to be signed by your local contact before an account can be created.

It is suggested that a record of your analysis is kept in a word document. This can be done using the procedures given in Appendix 8. This should include for example, names of files created, copies of plots you wish to keep and the mean values of relevant peaks areas and widths which are produced by the data analysis routines.

## **2. Viewing the raw time of flight data**

### **2a Obtaining raw time of flight data**

VESUVIO data is obtained using resonance foil methods [1,2,3,4,5] as outlined in Appendix 1. The “raw” data in each detector is a linear combination of time of flight spectra collected with the gold foils in different positions. Typically each experiment is divided into a series of identical consecutive runs each of 900  $\mu$ amps. The commands RAWB and RAW sum all runs between the first and last run number entered. If only one run is to be analyzed the first and last run numbers entered should be identical. If you wish to sum runs which are not consecutively numbered, the commands described in Appendix 7 should be used.

It is recommended that you view the raw data as early as possible to check that the run is proceeding in a satisfactory way. Possible reasons for non-satisfactory data are; the shutter is closed, the command file which moves the resonance foils to different positions is not running, the sample is not in the correct position etc.

To obtain the raw back scattering data, the “RAWB” command is used.

```
eVS> rawb
```

```
first and last run numbers?
```

```
14188 14195
```

```
first and last spectrum numbers?
```

```
3 134
```

This produces an output file with the name TEMPB.DAT, containing the time of flight data for each detector at back scattering. Many of the files produced by the analysis procedures are temporary files which should be deleted once the final data set has been obtained. Such files are given a name beginning with TEMP. Details of the format of all ASCII files produced by the analysis routines are given in Appendix 6.

To obtain the raw forward scattering data the “RAW” command is used.

```
eVS> raw
```

```
first and last run numbers?
```

```
14188 14195
```



first and last spectrum numbers?

135 182

This produces an output file with the name TEMP.DAT, containing the time of flight data for each detector at forward scattering angles.

## 2b Displaying data

The data in the files produced by RAW and RAWB can be displayed using the command PLOTD. This allows the display of single detectors, the sum of detectors within a specified angular range or the sum of consecutively numbered detectors For example;

eVS> plotd

Name of file containing time of flight data?

tempb

This is the filename produced by the RAWB routine, which contains the back-scattering data.

input file is tempb

tmin,tmax specify range which is plotted

tmin,tmax?

50 500

These are the limits of the time of flight range to be plotted. The range 50-500 includes all useful data at both back and forward scattering.

number of points= 900

ndmin= 3 ndmax= 134

plot individual detectors (1)

or sum of detectors within angular range (2)

or sum of consecutive detectors (3)

2

Enter minimum and maximum angles

150 170

The total range of back scattering angles is 130-170 degrees. See Appendix 1 and reference [3]. The plot produced by this input is of the data from detectors with angles between 150 and 170 degrees. To view the sum of all detectors enter 0 180 here.

```
detector    40  theta= 151.9331
detector    41  theta= 151.8561
detector    42  theta= 154.6456
detector    43  theta= 154.6228
detector    44  theta= 160.2408
```

.....

```
detector   131  theta= 154.7853
detector   132  theta= 157.3510
detector   133  theta= 160.8139
detector   134  theta= 163.5170
```

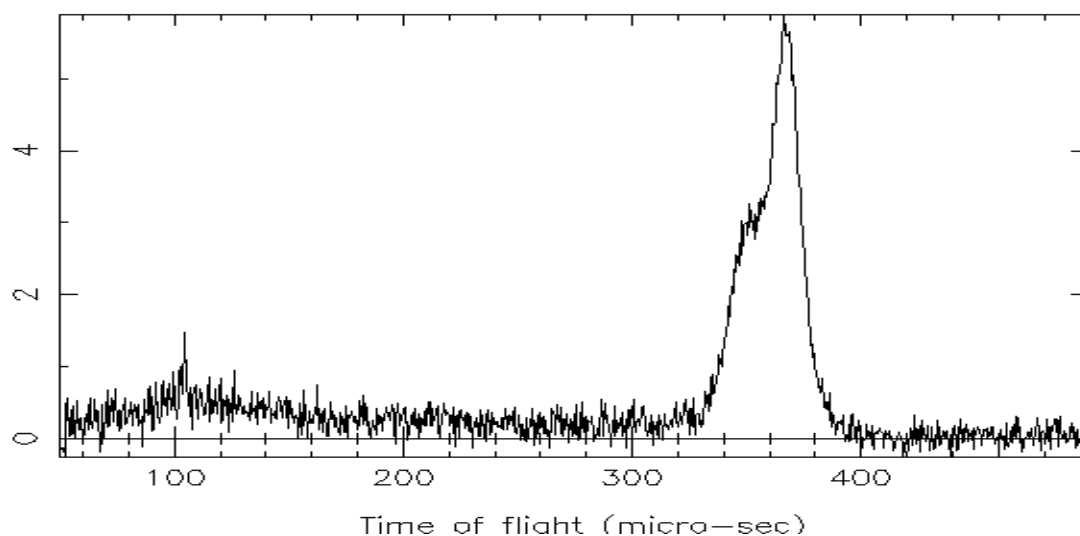


Figure 3. Plot produced by PLOTD. The sum of all detectors in the angular range 150-170° as specified in the input to PLOTD is shown. The non-zero background to the left of the Al/Zr peak is due to multiple scattering.

The program displays (Figure 3) the sum of all detectors with scattering angles between 150° and 170° between 150 and 500  $\mu$ sec. These limits are determined by the input from the user to PLOTD. PLOTD also lists the spectrum numbers which are included in the sum, with the corresponding scattering angles. This plot and any other plot you wish to keep can be pasted into a word document using procedure 2 given in Appendix 8.

The forward scattering data can be viewed in a similar way.

eVS> plotd

name of file containing input data?

temp

tmin,tmax?

50 562

This is the range of time of flight values which will be displayed

number of points= 1024

ndmin= 135 ndmax= 182

plot individual detectors (1)

or sum of detectors within angular range (2)

or sum of consecutive detectors (3)

enter 0 to exit

2

Enter minimum and maximum angles

60 80

detector 135 theta= 66.59930

detector 136 theta= 64.23130

detector 137 theta= 62.36170

detector 138 theta= 61.14130

```
detector    139 theta= 66.48230
detector    140 theta= 64.26080
detector    141 theta= 62.38260
detector    142 theta= 61.23530
```

The program prints out the detectors included with the scattering angles. Figure 4 shows the plot produced by PLOTD with the input above.

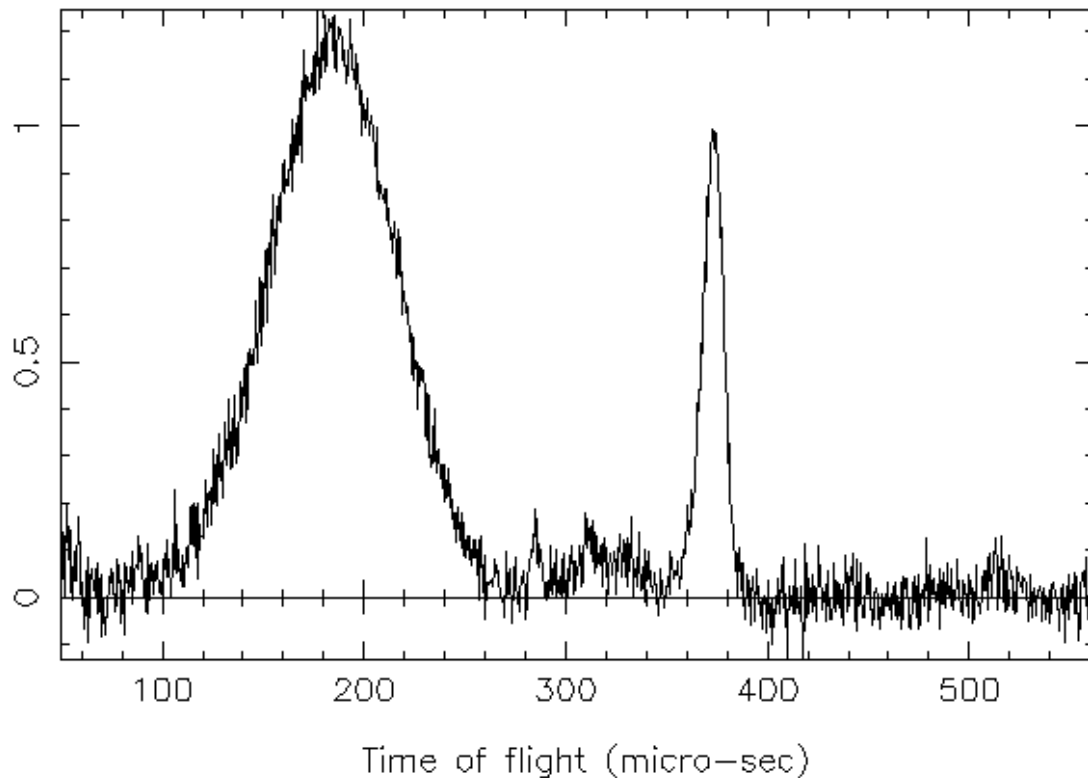


Figure 4. Shows the plot produced by PLOTD. It shows the sum of detectors with scattering angles between 60 and 80 degrees. The narrow peak at  $\sim 285 \mu\text{sec}$  is due to a nuclear resonance from Hafnium impurities. The broad feature between 300 and 350  $\mu\text{sec}$  is due to gamma background.

### 3. Correction of data for multiple scattering and gamma background.

For accurate results the data must be corrected for multiple scattering in the sample [6]. Multiple scattering from hydrogen can be observed as a non-zero background to the left of the peak from Al/Zr in Figure 3. The forward scattering data also contains a gamma background [2] which must be removed. The effects of this background can be seen in Figure 4, where it produces a broad peak between 300 and 370  $\mu\text{sec}$ . Both of these effects are determined by the scattering properties of the sample + container. These corrections are performed automatically by the procedure given in this section. A more detailed account of the correction procedures in this section is given in Appendix 10.

To perform the corrections you need to know;

- (a) The atomic masses present in your sample and container

(b) The neutron transmission of the sample and sample can.

In order to determine the sample+can transmission it is necessary to perform an “empty beam run” with no sample or can in the beam. This run should be performed at the start of your experiment. Only a short run of ~30 minutes is required, The sample transmission is obtained using the command **TRANS** and two runs; the empty beam run and a run with the sample+can in the beam.

```
eVS> trans
```

```
ENTER SAMPLE RUN NUMBER
```

```
14188
```

```
ENTER CAN/EMPTY BEAM RUN NUMBER
```

```
14187
```

```
sample transmission= 0.8316979
```

```
sample attenuation= 0.1683021
```

The TRANS program calculates the sample+can transmission by taking the ratio of counts in the incident beam monitor S1 (see figure A1.1, P41) and the transmitted beam monitor S2.

The next step is to create a command file for data correction using the command CRECOM. First you need to determine the name of the directory in which you are working.

```
eVS> sh def
```

```
USER$DISK:[JM01]
```

Then type “CRECOM”

```
eVS> crecom
```

```
Directory in which you are working?
```

```
Type sh def before starting program to determine this
```

```
USER$DISK:[JM01]
```

```
Generic file name?
```

```
zh
```

The “Generic file name is chosen to indicate the sample and experimental conditions. It can be up to 16 characters in length.

```
There are 3 options
```

```
(1) A single run
```

```
(2) A number of consecutively numbered runs
```

```
(3) A number of non-consecutive runs
```

```
Enter option 1,2,or 3
```

```
2
```

```
First and last run numbers?
```

```
14188 14195
```

```
Number of different atomic masses (maximum=7) in sample+can
```

3

A maximum of 7 masses can be included.

Enter mass 1

1.0079

This is the mass in amu of the hydrogen atoms in the sample.

Enter mass 2

27

This is the mass of the aluminium atoms in the sample container

Enter mass 3

91

This is the mass of the zirconium atoms in the sample.

Is sample at  $T < 70\text{K}$ ? (Y=1,N=0)

0

The correction and fitting procedures are slightly different for high and low temperatures due to the presence of “Final State Effects” (See Appendix 4). In this case the sample was at room temperature.

Does sample contain H (1) or D (2) or D+H (3)

1

There are 3 options (1) H only, (2) D only, (3) Both D and H (for example when partially deuterated samples are run). The sample contains only H in this case.

Fast (1) or slow (2) option?

1

The procedure calculates the multiple scattering by a Monte-Carlo integration [6]. The fast option uses 10 times fewer events than the slow option and is usually adequate for most samples. If the fast option is chosen the correction procedure runs in ~10 minutes. The slow option takes ~90 minutes.<sup>1</sup>

Transmission of sample+can?

0.831

This is the value determined using the TRANS command at the start of this section.

enter tmin,tmax for data to be excluded

to exit enter tmin=tmax

278 292

---

<sup>1</sup> If you wish to check whether your data is sensitive to the accuracy of the MS correction the slow option can be chosen and the procedure re-run. Alternatively different runs of CRECOM will give slightly different results due to the randomness in the MS calculation. Hence the sensitivity of the results to the accuracy of the MS calculation can also be tested by rerunning ZH.COM on the fast option.

This option allows regions of data to be excluded. In this case there is a spurious peak at ~285  $\mu$ sec which is produced by a neutron resonance from Hafnium impurities in the ZrH<sub>2</sub> (see Figure 4). The region containing this peak is therefore excluded. If you are unsure of whether or not to eliminate data type "0 0" here.

```
enter tmin,tmax for data to be excluded
to exit enter tmin=tmax
0 0
```

The procedure above produces a file ZH.COM. This is reproduced in Appendix 9. If your "generic file name" was XXXX this will be XXXX.COM. Throughout this guide the generic file name ZH is used – hence ZH should be replaced by your chosen generic file name in analysis of your data. The file ZH.COM can be run interactively by the command

```
eVS> @zh
```

It can also be run as a batch job using the following procedure.

```
eVS> sh def
USER$DISK:[JM01]
eVS> submith
ENTER FILENAME: zh
ENTER DIRECTORY: USER$DISK:[JM01]
Job ZH (queue HATHOR$SLOW, entry 2746) started on HATHOR$SLOW
%DCL-W-SKPDAT, image data (records not beginning with "$") ignored
```

To check that the job is running, type "q"

```
eVS> q
Batch queue HATHOR$SLOW, busy, on HATHOR::
```

Entry	Jobname	Username	Status
2746	ZH	JM	Executing

When the procedure has run (this takes 10-15 minutes if the fast option has been chosen), the corrected data for forward scattering is contained within the file ZH.DAT and the back scattering data is in ZHB.DAT. The data can be displayed using PLOTD. For example;

```
eVS> plotd
Name of file containing time of flight data?
zh
input file is zh
```

```
tmin,tmax specify range which is plotted
tmin,tmax?
50 562
```

```

number of points=    1024
ndmin=    135 ndmax=    182
plot individual detectors (1)
or sum of detectors within angular range (2)
or sum of consecutive detectors (3)
enter 0 to exit

```

2

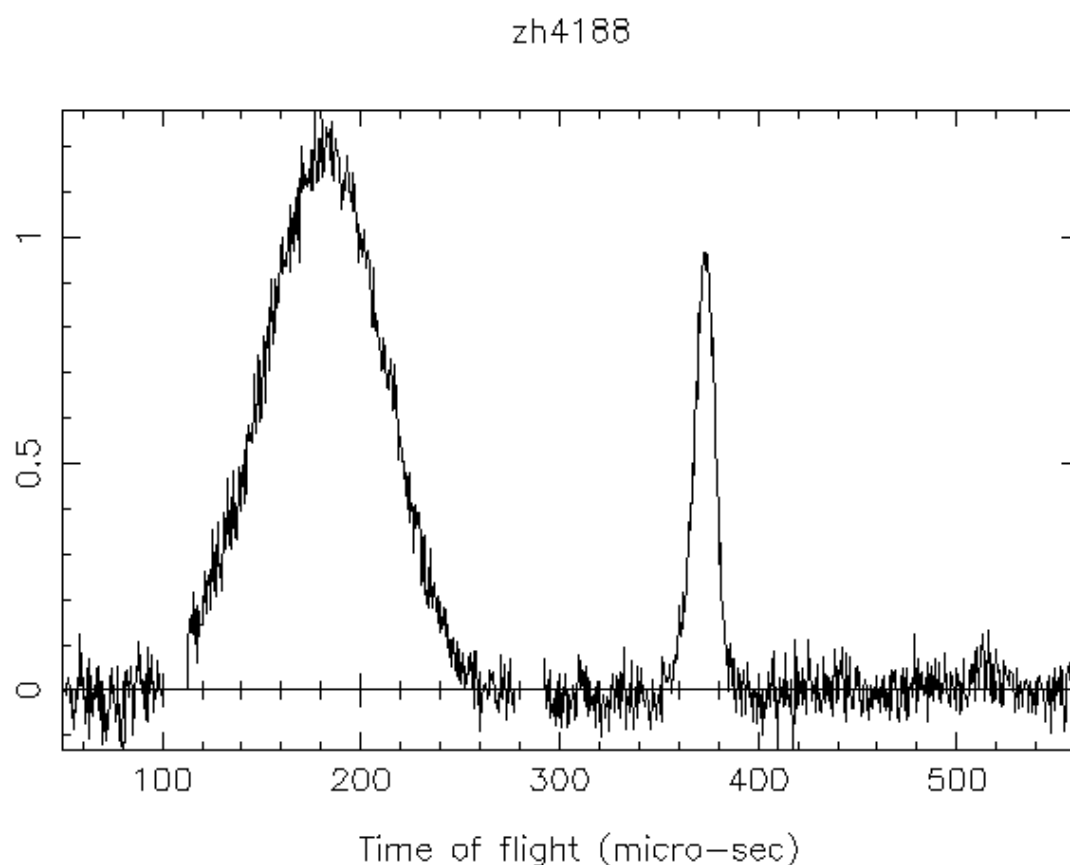


Figure 5 Shows the sum of data with angles between 60 and 80 degrees after the correction procedure performed by ZH.COM.

Figure 5 shows the sum of data with angles between 60 and 80 degrees after the correction procedure performed by ZH.COM. The procedure automatically removes data in the region 100-120  $\mu\text{sec}$  since this region contains a contribution from a second absorption in the gold analyser foil at  $\sim 60$  eV. The region 272-292  $\mu\text{sec}$  was excluded in response to the user input to the CRECOM routine on page 11. These regions are given very large error bars so that they do not affect any of the fitting procedures. It can be seen that the broad feature between 300 and 350  $\mu\text{sec}$  seen in Figure 4 has been removed by the correction procedure.

If you wish to understand in more detail the procedures used by the CRECOM command it is suggested that you use the CRECOM2 rather than the CRECOM command. The former

command produce an identical file (ZH.COM) to CRECOM except that the final line which deletes all intermediate files is omitted. Hence after the procedure has run, your area will contain a number of intermediate files which can be examined. These are;

1. (a) TEMP.DAT containing the “raw” data from forward scattering  
(b) TEMPB.DAT containing the “raw” data from back scattering.
2. (a) TEMPD.DAT containing data and fits from TFIT on TEMP.DAT  
(b) TEMPP.DAT contains parameters from fit to TEMP.DAT  
(c) TEMPBD.DAT containing data and fits from TFIT on TEMPB.DAT  
(b) TEMPBP.DAT contains parameters to fit to TEMPB.DAT
3. (a) TEMPC.DAT – gamma corrected forward scattering data  
(b) TEMPCD.DAT. TEMPCP.DAT data and fits and parameters from fit to TEMPC.
4. Files ZH91.dat – ZH182.DAT containing the multiple scattering data for each detector produced by the MSCALC routine
5. (a) TEMPCMS – forward scattering data corrected for gamma background and multiple scattering.  
(b) TEMPCMSP.DAT and TEMPCMSD.DAT – parameters, data and fits to TEMPCMS  
(c) TEMPBMS – back scattering data corrected for gamma background and multiple scattering.  
(d) TEMPBMSP.DAT and TEMPBMSP.DAT – parameters, data and fits to TEMPBMS.DAT.
6. Files ZH.IN, ZHB.IN – these are input files to TFIT for forward and backscattering.  
ZHMS.IN – the input file to MSCALC for calculation of multiple scattering.

These files can be examined using the routines given in the next section (PARMEAN, TFITPLOT). In practice the most unreliable part of the automatic correction procedure is the multiple scattering correction. It is suggested that this is examined using the routine MSSUB (see section A10.4). All the commands used by the CRECOM procedure are detailed in Appendix A10.

The intermediate files use considerable disk space and should be deleted once you are satisfied that the correction procedure has worked satisfactorily. This can be done using the command

eVS> **delfiles**

#### **4. Fitting the data to obtain sample compositions and kinetic energies.**

The simplest physical analysis which can be made of the data is to assume that all the atomic momentum distributions have a Gaussian form in momentum space (see Appendix 2). The command TFIT fits the time of flight data from each detector with this assumption. The fitting function used is described in detail in Appendix 2. The output of TFIT contains the Gaussian



widths and the amplitudes of all peaks which are included in the fit. The values of the fit parameters obtained from TFIT can be used to determine the atomic kinetic energies and the sample composition.

#### 4a Back Scattering

The widths and amplitudes of the peaks in the data are obtained using the TFIT command. This requires an input file containing the masses of all atoms with mass greater than 3. The file ZHB.in for fitting the back-scattering data is produced automatically by running the command file ZH.COM produced by the CRECOM command (see section 3). Alternatively this file can be produced using the command CREBIN (create backscattering .in file) .

```
eVS> crebin
```

```
Generic file name? (must be < 16 characters)
```

```
ZH
```

```
number of masses in the sample+can other than M=1 or M=2
```

```
2
```

```
mass      1
```

```
27
```

```
mass      2
```

```
91
```

```
Was the sample at T<70K? (Yes=1,No=0)
```

```
0
```

This sequence of inputs creates a file ZHB.IN of the following form (if your generic file name was XXXX, a file XXXXB.IN would be produced),

```
2 ! 1 = forward scattering (135-182). 2 = back scattering (3-134)
```

```
2 !Number of masses in sample
```

```
27.0000 0 0 !Mass of atom, amplitude, width
```

```
91.0000 0 0 !Mass of atom, amplitude, width
```

```
0 ! FSE are not fitted
```

The lines in ZHB.IN have the following functions.

1. Option 1 is forward scattering. Option 2 is back scattering. Option 3 is back-scattering for single difference data.
2. The number of masses in the sample+container. In this case there are only two masses since no H peak appears at back-scattering. TFIT allows up to seven different masses to be fitted.
3. The lightest mass in amu. In this case this is the aluminium of the sample container. The two zeros following imply that both the widths and the amplitude of the peak are fitted. If either of these parameters is non-zero the width or amplitude is fixed at the value entered (see sections 4d and 4f for an example of this procedure).
4. As for line 3 but for the Zirconium in the sample

5. If this number is 1, a "Final State" correction is made (see Appendix 3). This is required for fitting H and D peaks and for data at low temperatures. It is usually set to zero for analysis of back-scattering data at room temperature. It should be set to one if data at temperatures less than ~70K are used.

The text after the exclamation mark (!) is a comment which is not read by the programs.

This file can be edited by hand using the command

```
eVS> nedit zhb.in
```

This allows amplitudes or widths of particular peaks to be fixed or the inclusion of more atomic masses.

TFIT fits data between user specified limits  $t_{\min}$  and  $t_{\max}$  in the time of flight spectra. These limits are chosen to cover the data region of interest. Due to the incorporation of fast Fourier transforms in the programs,  $t_{\max} - t_{\min}$  must be a power of 2.

To fit the data type TFIT.

```
eVS> tfit
```

```
Name of file containing time of flight data?
```

```
Zhb
```

```
input file is zhb
```

```
tmin,tmax? (tmax-tmin must be 2**n)
```

```
300 428
```

This range was chosen by observation of the data shown in Figure 2. It includes the peaks from atoms other than hydrogen in the data, For most samples 300-428 is a good range to choose. For samples containing masses <7 the range may have to be extended to include all peaks: for example 200-456  $\mu$ sec.

```
number of points= 256
```

```
ndmin= 3 ndmax= 134
```

```
Name of file containing fit parameters?
```

```
Zhb.in
```

```
npeaks= 2
```

```
1 am= 27.00000 xs= 0.0000000E+00 wid= 0.0000000E+00
```

```
2 am= 91.00000 xs= 0.0000000E+00 wid= 0.0000000E+00
```

```
FSE NOT SUBTRACTED
```

```
First and last spectrum number?
```

```
3 134
```

The routine fits the data in spectra 3-134 and produces two output files. These files have the same name as the input data file with the letter D or P added. In the example these are;

(1) ZHBD.DAT- This contains the data and fits

(2) ZHBP.DAT – this contains the fitted parameters.

If your generic file name was XXXX, these will be XXXXBD.DAT and XXXXBP.DAT. These are ASCII files with formats which are given in Appendix 6.

#### 4b. Displaying the fits

The data and fits can be displayed using the TFITPLOT command. This allows one to plot the data, fit and individual peak components to the fit. This can be done either for individual spectra, the sum of the spectra within a specified angular range or for consecutively numbered detectors.

```
eVS> tfitplot
name of file containing input data?
Zhbd
plot individual detectors (1)
plot sum of detectors between set angles (2)
plot sum of consecutive detectors (3)
2
enter minimum and maximum angles
150 170
40 th= 151.9331
41 th= 151.8561
.....
133 th= 160.8139
134 th= 163.5170
```

The program lists the detectors included. It sums the data and fits from all these detectors and plots the data, fit and the individual peaks contributing to the fit in the time of flight range chosen in the input of TFIT.

The plot produced by the input above is shown in Figure 6. The black line is the sum of data from spectra with angles between the input limits of 150 and 170 degrees. The red line is the fit. The blue line is the fitted contribution of the Zr peak and the green line the fitted contribution of the Al container.

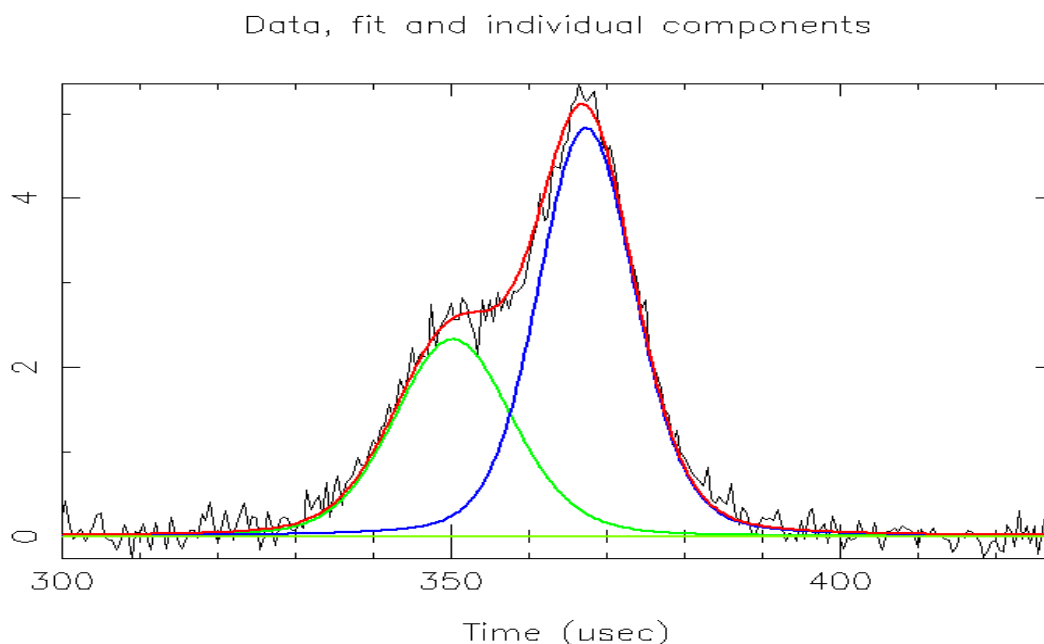


Figure 6. Data and fit given by TFIT for detectors with scattering angles between 150 and 170 degrees. The data is black), the fit is red and the individual peaks contributing to the fit are green (aluminium) and blue (zirconium).

#### 4c. Examining Fitted Parameters

A quick overview of the parameters can be obtained using the command PARMEAN. This calculates;

- (a) The mean of the fitted parameters, weighted by the statistical errors on these parameters.
- (b) The unweighted mean and the standard error in the mean.

If the instrument and the analysis programs were perfect the only errors would be statistical. The two means (a) and (b) would then give the same results within error. In practice (b) always gives larger errors than (a). This is due for example to; faulty detectors, background effects, failures of the fitting program to find the global minimum in parameter space, inaccuracies in the assumptions used in the data analysis (for example that the momentum distribution is Gaussian) etc. As a rule of thumb, if absolute values are required the error should be taken as the standard error in the mean. However if, for example, only the sample temperature changes between runs, the statistical error becomes relevant, since one would expect systematic errors such as those just listed to be the same for all runs.

In the example;

```
eVS> parmean
```

```
Name of file containing output of TFIT routines?
```

```
tempbp
```

```
Number of masses=      2
```

```
first spectrum=      3 last spectrum=    134
```

1 ATOMIC MASS= 27.00000

Number of points included= 117  
wtd mean area= 0.4108056 +- 2.8046921E-03  
mean area = 0.4232959 st dev= 4.4313911E-03

Number of points included= 124  
wtd mean width= 12.78885 +- 0.2786912  
mean width = 14.40658 st dev= 0.3265285

2 ATOMIC MASS= 91.00000

Number of points included= 117  
wtd mean area= 0.5891944 +- 2.8046921E-03  
mean area = 0.5767043 st dev= 4.4313911E-03

Number of points included= 124  
wtd mean width= 26.62335 +- 0.2796181  
mean width = 26.80821 st dev= 0.4397886

2

27.00000	0.4108056	12.78885
91.00000	0.5891944	26.62335

The program PARMEAN automatically eliminates outliers from the averages;

- (1) The mean value  $\chi^2_m$  of the reduced  $\chi^2$  of the fit to each detector is calculated and detectors giving fits with  $\chi^2 > 1.2 \chi^2_m$  are excluded. Thus only detectors with good fits contribute to the mean.
- (2) The mean value of each parameter over all detectors is calculated. Points differing from the mean by more than 3 times the counting error on the point are excluded. This eliminates outliers due for example to faulty detectors.
- (3) Points with very large error bars are excluded (more than 4 times the value of the point). This again eliminates faulty detectors.
- (4) Points with very small error bars are also excluded. The fitting routine occasionally produces anomalous values with very small error bars. This is probably due to the minimisation routine getting stuck in a local minimum in parameter space.

Consequently, as specified in the output given above, the number of points included in the calculation of the mean values is less than the total number of detectors. For example in the calculation of the mean width of the Zr peak above, only 124 of the 134 back scattering detectors were included in the calculation of the mean.

The final 3 lines of the output contain a summary of the results; the number of masses, and the mass, mean amplitude and mean width for each mass. A more detailed examination of the fitted areas and widths can be made by use of the routine PARPLOT.

eVS> **parplot**

Name of file containing output of TFIT routines?

**tempbp**

Number of masses= 2

NUMBER OF DETECTORS= 132

The file contains data on the following masses

1 M= 27.0000

2 M= 91.0000

Examine data on peaks areas (1) or widths (2)?

Type 0 to exit program

If 2 is entered in response to this prompt the fitted peak widths are displayed.

**2**

Examine peak widths.

Mass number?

**1**

PEAK NUMBER 1 MASS= 27

Arrange in order of increasing angle (Y=1,N=0)?

**1**

If 0 is entered in response to the above prompt the widths are displayed as a function of detector number rather than angle.

	ANGLE	Y	E
3	1.3047E+02	1.5353E+01	4.8343E+00
4	1.3193E+02	1.3269E+01	5.6182E+00
5	1.3306E+02	1.5290E+01	3.6656E+00
6	1.3280E+02	2.1783E+01	1.1835E+01
.....			
88	1.5733E+02	1.3529E+01	1.9359E+00
89	1.6034E+02	1.4563E+01	2.2009E+00
90	1.6313E+02	1.0277E+01	1.6937E+00

Plot widths? (Y=1,N=0)

The program prints out the detector numbers, the corresponding scattering angle, the value of the parameter plotted (width in this case) and the statistical error on the value.

Plot widths? (Y=1,N=0)

**1**

MEAN VALUE GIVEN BY PARMEAN= 11.73119 +- 0.3828053

This is shown as the red line

The program calculates the mean value given by PARMEAN for comparison with the data and displays it as a red line.

Graphics device/type (? to see list, default /NULL):/XW

The plot can be displayed on the screen by /XW. It can be written to a .gif file by /GIF and to a .ps file by /PS. The plot produced by the sequence of commands followed is shown in Figure 7.

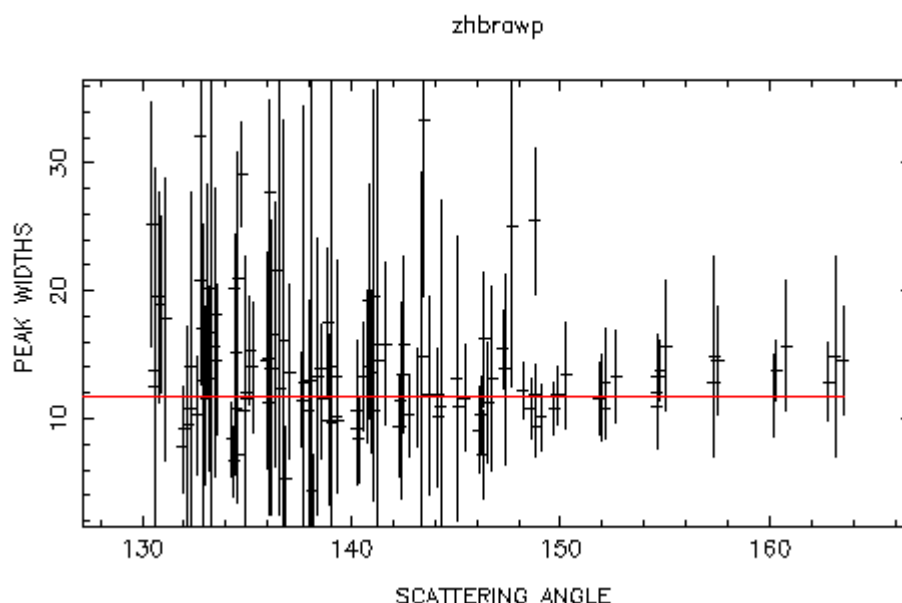


Figure 7. Widths for peak 1 (aluminium) obtained with the double difference data at back-scattering in spectra 3-90. The red line is the mean value given by PARMEAN.

CHANGE LIMITS? (Y=1,N=0)

0

If 1 is entered here, the program prompts for new x and y limits on the plot and plots again with the new limits. Otherwise the program proceeds to the calculation of mean values.

READ BAD DETECTORS FROM FILE (1) OR TERMINAL (2)

IF READING FROM FILE, FILE "EXCLUDE.DAT"

MUST HAVE BEEN BE PREPARED

2

The program allows the elimination of unreliable detectors.

NUMBER OF DETECTOR TO BE EXCLUDED (0 TO END)

55

0

The program gives the option of entering unreliable detectors by hand or reading from a file "exclude.dat". This saves keyboard strokes if many data sets are analysed and some

detectors are known to be unreliable. If option 2 is chosen the file EXCLUDE.DAT must be present in the directory in which you are working. This can be created by hand using the NEDIT or EDIT command and must have been prepared before PARPLOT is run. It contains the same input as would be read by hand if option 2 was entered in response to the prompt. The input above would exclude detector 55 from the mean value calculation. A file EXCLUDE.DAT of the form

55

0

Would also eliminate detector 55 from the mean. The file exclude.dat can be prepared using either the ed, (ctrl z to exit) ed/edt (ctrl z followed by ex to exit) or nedit commands (exit using menu at top of window).

The following detectors are excluded from averages

55

Calculate mean value of width

CALCULATE MEANS

FIRST AND LAST POINTS?

TO EXIT PUT FIRST POINT=LAST POINT

3 134

3	XV=	130.4653	X=	15.35300	+ -	4.834300
4	XV=	130.6171	X=	13.71800	+ -	15.92800
5	XV=	130.6221	X=	12.55400	+ -	12.51400

.....

132	XV=	162.8193	X=	12.89400	+ -	3.085300
133	XV=	163.1337	X=	14.84800	+ -	7.824900
134	XV=	163.5170	X=	14.54000	+ -	4.164900

WTD MEAN= 11.93843 + - 0.3689854

MEAN= 13.77069 ST.DEV= 0.4116524

The program prints out the detectors included in the average, in order of increasing angle in response to the choice of option 1 on page 19. It calculates the weighted mean, taking into account the statistical errors on the fitted widths. It also calculates the mean of the values and the standard error in the mean.

FIRST AND LAST POINTS?

TO EXIT PUT FIRST POINT=LAST POINT

Means of selected ranges can be taken by entering appropriate numbers. For example

76 79

76	XV=	149.8588	X=	13.72800	+ -	2.974500
77	XV=	151.8561	X=	13.51600	+ -	4.587000



```

78 XV= 151.8824   X= 15.13600   +- 2.984200
79 XV= 151.9331   X= 12.64900   +- 1.704400
WTD MEAN= 13.36597   +- 1.273004
MEAN= 13.75725   ST.DEV= 0.5154468

```

calculates the means of detectors 76-79 with scattering angles 149.8 - 151.9 degrees.

FIRST AND LAST POINTS?

TO EXIT PUT FIRST POINT=LAST POINT

0 0

The program returns to the initial prompt if first point=last point.

The file contains data on the following masses

1 M= 27.0000

2 M= 91.0000

Examine data on peaks areas (1) or widths (2)?

Type 0 to exit program

0

Similar analysis and plotting can be made of other fitted parameters – e.g. widths of different masses or amplitudes instead of widths.

#### 4d Forward Scattering

The forward scattering data is fitted in a similar way. Usually the widths and amplitudes of the atoms with  $M > 3$  are fixed at the values determined from the back scattering data, since the latter data gives more accurate values for heavier masses. The File ZH.COM created in section 3 using the CRECOM procedure automatically produces a file ZH.IN with the values for heavier atoms fixed at the values produced from fitting the back-scattering data.

Alternatively this file can be created using the CREFIN (create F.IN file) command.

eVS> crefin

Name of file containing output of TFIT routines?

Zhbp

Number of masses= 2

first spectrum= 3 last spectrum= 134

1 ATOMIC MASS= 27.00000

Number of points included= 122

.....

wtd mean width= 26.75393 +- 0.2900341

mean width = 26.87679 st dev= 0.4481016

Enter generic file name

Zh

mass 1 or 2?

1

This creates a file ZH.IN of the following form.

```
1      ! Option. (1) Forward scattering
      3
1.0079 0 0
27.00000  0.4223358  13.13998
91.00000  0.5776640  26.64120
1      ! NFSE
```

Comparing this file to ZHB.IN, it can be seen that the widths and amplitudes of the peaks from masses 27 and 91 are fixed at the values obtained from back-scattering. The TFIT program is written in such a way that only the ratio of the fixed amplitudes is significant (that is the same results would be produced if 0.4223358 was changed to 2 x 0.4223358 and 0.5776640 to 2 x 0.5776640). Again this file can be modified by hand using the NEDIT command if you want to try different constraints on the fitting.

The forward scattering data set ZH.DAT was fitted, using the input file ZH.IN

```
eVS> TFIT
```

```
Name of file containing time of flight data?
```

```
Zh
```

```
tmin,tmax? (tmax-tmin must be 2**n)
```

```
50 562
```

```
Name of file containing fit parameters?
```

```
Zh.in
```

```
input file is zh
```

```
number of points= 1024
```

```
ndmin= 135 ndmax= 182
```

```
First and last spectrum number?
```

```
135 182
```

The routine produces two output files; ZHD.DAT which contains the data and fits for each detector and ZHP.DAT, which contains the fitted parameters. The fits can be examined using TFITPLOT and the input files ZHD.dat. The fitted parameters in zhp.dat can be examined using PARMEAN and PARPLOT as described in section 4c.

The command **PARPLOT** gave the plot shown in Figure 8 for the hydrogen peak widths.

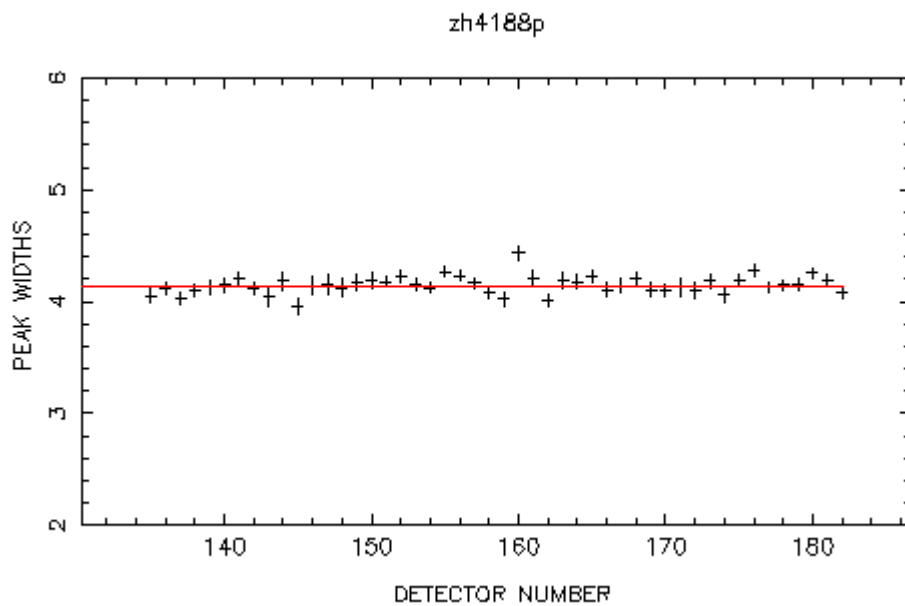


Figure 8. Values of the H peak width as a function of detector number after corrections for gamma background and multiple scattering.

...

The hydrogen peak areas obtained from the corrected data are shown in Figure 9

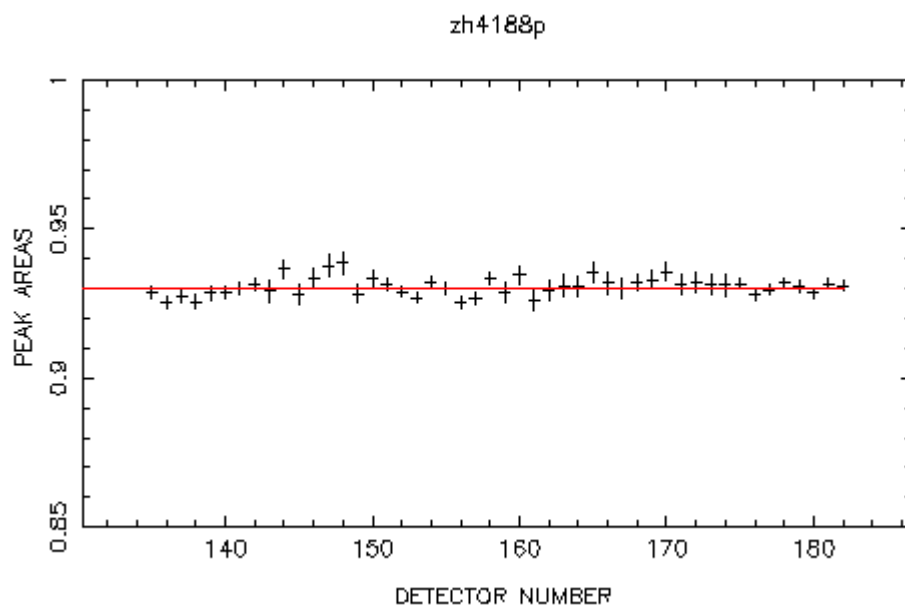


Figure 9. Values of the H peak areas obtained from the fits as a function of detector number.

The mean values of the widths and areas were obtained using the command **PARMEAN**.

**eVS> parmean**

Name of file containing output of TFIT routines?

**zh**

Number of masses= 3

first spectrum= 135 last spectrum= 182

1 ATOMIC MASS= 1.007900

Number of points included= 48

wtd mean area= 0.9292901 +- 3.3597506E-04

mean area = 0.9298095 st dev= 4.4459224E-04

Number of points included= 46

wtd mean width= 4.137728 +- 8.1897276E-03

mean width = 4.132648 st dev= 1.1147558E-02

2 ATOMIC MASS= 27.00000

Number of points included= 47

wtd mean area= 2.6493629E-02 +- 1.2845309E-04

mean area = 2.6321936E-02 st dev= 1.6461386E-04

Number of points included= 48

wtd mean width= 11.50000 +- 1.4433755E-07

mean width = 11.50000 st dev= 0.0000000E+00

3 ATOMIC MASS= 91.00000

Number of points included= 47

wtd mean area= 4.4036966E-02 +- 2.1351091E-04

mean area = 4.3751560E-02 st dev= 2.7361230E-04

Number of points included= 47

wtd mean width= 26.60000 +- 5.7735025E-07

mean width = 26.60000 st dev= 0.0000000E+00

#### 4e Calculation of kinetic energies, vibration frequencies and sample composition

The widths obtained from PARMEAN can be converted to kinetic energies or equivalent harmonic oscillator frequencies using the command KE. For example the hydrogen width;

eVS> ke

ENTER MASS (AMU)

1.0079

ENTER MEASURED WIDTH AND ERROR (A-1)

4.137 0.011

This is the value and error given by PARMEAN for the H peak width.

Measured profile width= 4.137000 +- 1.1000000E-02 A-1  
 Kinetic energy= 107.3188 +- 0.5707068 meV  
 Kinetic energy= 1245.328 +- 6.622482 K  
 Fundamental frequency= 143.0918 +- 0.7609425 meV  
 Fundamental frequency= 1154.107 +- 6.137382 cm-1

The KE program calculates the kinetic energy of the atom in meV. The kinetic energy is also given in degrees. Note that for hydrogen this is a very high temperature. At room temperature the kinetic energy of protons and deuterons is almost entirely due to their quantum zero-point motion. For heavier masses the temperature of the sample makes a significant difference to the kinetic energy of the atoms.

The KE program also gives the “fundamental frequency” in meV and in  $\text{cm}^{-1}$ . This is the frequency of a 3D isotropic harmonic oscillator which would give the same Gaussian width to the momentum distribution as that fitted. ZrH<sub>2</sub> closely approximates an isotropic harmonic oscillator and thus the value of fundamental frequency obtained in the example above is very close to the values measured by neutron spectroscopy [27]. These lie in the range 141-147 meV.

The mean of the fitted values of peak areas can be used to determine the relative numbers of different atoms in the sample+can. For example the values of the peak areas determined by PARMEAN is listed in the second column of table 1. The neutron cross-sections for the elements are listed in the table on the wall to the right of the control terminal in the VESUVIO cabin. They can also be readily obtained from the internet. These are given in column 2 in barns for the elements in the sample+can. The number of atoms in the beam are in the same ratio as the fitted area divided by the cross-section, given in column 3. The results imply that the number of atoms in the beam was in the ratio 1.137 (H):1.75 (Al):0.67 (Zr)

Mass	Peak area	Cross-section	Area/cross-section
1.0079	0.9292 (4)	81.67	0.01137
27.0	0.0264(1)	1.506	0.0175
91.0	0.0440 (2)	6.56	0.0067

Table 1 Peak areas and cross section for ZrH<sub>2</sub> + al can.

**Once you are satisfied with the quality of the fits and have copied any plots you require , the files used by TFITPLOT should be deleted as they use much disk space.**

eVS> delete \*d.dat;\*

**These files can be rapidly regenerated if necessary by running TFIT.**

#### 4f. Example 2 Back scattering

When the sample composition is known, the amplitudes of the peaks from atomic masses in the sample can be fixed. This allows more accurate determination of atomic kinetic energies. The sample used here to illustrate this procedure contained O, Na, Si, Cu in known concentrations. The back scattering bank was used as this gives the best separation of the separate peaks. The sample signal overlapped with a strong aluminium signal from the sample container and the CCR used to cool the sample. Such signals can be subtracted using the command CANSUB.

CANSUB can also be run on forward scattering data, but this is not usually necessary for the study of H or D peaks since these are generally well separated from the can signal in the time of flight spectra. In order to run CANSUB it is necessary to know the sample transmission. The scattering from the can/sample environment depends upon this as the neutron beam is attenuated as it travels through the sample. For example the scattering from the front of the can is not the same as that from the back, since the neutron beam loses intensity as it passes through the sample. The sample transmission can be determined using the command **TRANS** as in section 3.

```
eVS> trans
```

```
ENTER SAMPLE RUN NUMBER
```

Here the sample+can run is entered

```
ENTER CAN/EMPTY BEAM RUN NUMBER
```

Here a run number with the empty can is entered. **Note that an empty can run must be used here and not the empty beam run used in section 3.**

```
sample transmission= 0.986034
```

Before CANSUB can be run, corrected data files containing the sample and empty can data must be produced using the procedure described in section 3. The sample run must be processed including all masses in the sample+container in the input to CRECOM. For processing the can run only the masses present in the can are required. In this example the corrected data was contained in CULTAB.DAT and CANB.DAT. The can subtraction was performed by the following procedure.

```
eVS> cansub
```

```
Name of file containing sample+can data?
```

```
cultab
```

```
File containing sample+can data is cultab
```

```
Name of file containing can data?
```

```
canb
```

```
File containing sample+can data is canb
```

Sample transmission?

.986

This is the value obtained above from TRANS on the (sample+can)/(empty can)

Name of output file ?

cultabs

Output file is cultabs

CANSUB plots the data before can subtraction (black), the data after subtraction (green) and the can signal subtracted (red) in user selected angular ranges.

Display input and output data

Enter thmin,thmax

150 180

40 thv= 151.9331

41 thv= 151.8561

.....

132 thv= 157.3510

133 thv= 160.8139

134 thv= 163.5170

The program lists the detectors included in the plot.

Black=sample+can. Red=can. Green=sample.

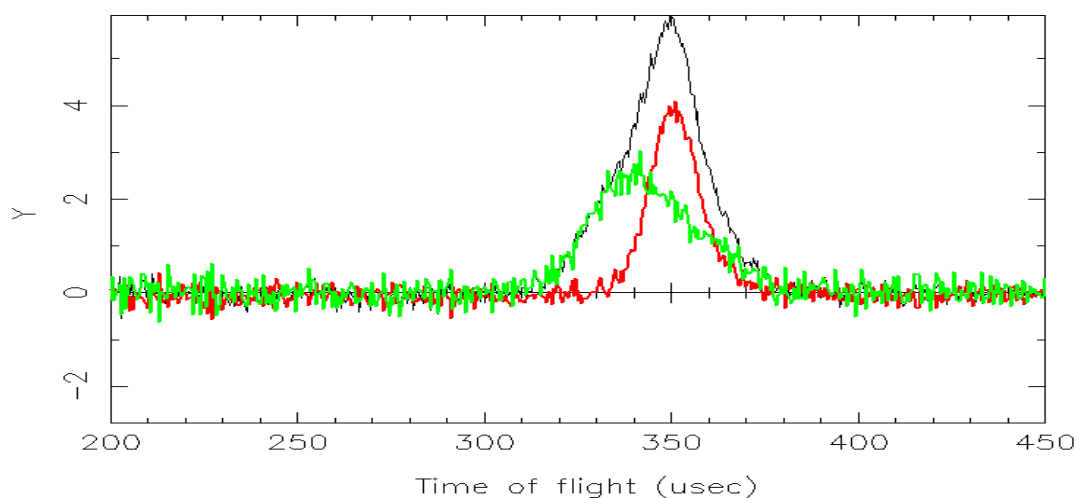


Figure 10. shows the data in the selected range 150-180 degrees before the can subtraction as the black line. The can signal is the red line. The data after the can subtraction is the green line.

Type <RETURN> for next page:

Enter thmin,thmax

0 0

The data after the can subtraction is now contained in the output file name specified in the input to CANSUB (CULTABS.DAT in this case)

The data after can subtraction can be fitted using the program TFIT as described in section 4a. In this case the sample composition was known and is listed in the table below.

Mass	no of atoms	Neutron xsection	fixed amplitude
16	6.81	4.23 barns	28.8
23	1.0	3.23 barns	3.23
28	1.74	2.17 barns	3.77
63.5	0.556	7.93 barns	4.41

The second column gives the ratio of the number of atoms mass M to the number of sodium atoms. Thus for example there are 6.81 oxygen atoms for each sodium atom. The fixed amplitude in the .in file for TFIT, given in column 4 is simply the number of atoms x the neutron scattering cross-section in barns (column 3). TFIT is written in such a way that only the ratio of these fixed amplitudes is significant

Thus the file CULTAB.IN shown below was created by hand, using the NEDIT command, The amplitude parameters were fixed at the values calculated from the table above

```

2          ! back scattering option
4          ! Number of masses in sample
16 28.8 0
23.0 3.23 0
28.0 3.77 0    ! Mass of atom, no of atoms x cross-section,width.If zero fit.
63.5 4.41 0
0          ! No final state subtraction

```

The data was then fitted using the following sequence of commands.

```

eVS> tfit
Name of file containing time of flight data?
cultabs
input file is cultabs

tmin,tmax? (tmax-tmin must be 2**n)
300 428
number of points=    256
ndmin=    3 ndmax=    134
Name of file containing fit parameters?
cultab.in
e1= 4897.400
de1g= 88.00000 de1l= 41.00000
ipno=    4

```



```

npeaks=      4
  1 am= 16.00000  xs= 28.80000  wid= 0.0000000E+00
  2 am= 23.00000  xs= 3.230000  wid= 0.0000000E+00
  3 am= 28.00000  xs= 3.770000  wid= 0.0000000E+00
  4 am= 63.50000  xs= 4.410000  wid= 0.0000000E+00

```

First and last spectrum number?

3 134

The fitted parameters are contained in the file cultabsp.dat and the data and fits in the file cultabsd.dat. These can be examined using TFITPLOT, PARPLOT and PARMEAN as described in section 4.

## 5. Determination of momentum distributions

The momentum distribution of atoms has a strictly Gaussian shape only if the atoms sits in an isotropic harmonic potential. The detailed shape of the momentum distribution  $n(p)$  is of interest as it contains information on the anisotropy of the binding and anharmonic effects. The procedures described here to obtain the shape of  $n(p)$  are illustrated by the determination of a proton momentum distribution. However they are equally valid for any data set in which the peak from one of the atomic masses is well separated from peaks due to other masses; for example D,  $^3\text{He}$ ,  $^4\text{He}$ , Li.

### 5a Isolation of the signal from hydrogen

The starting point of this analysis is the corrected time of flight data obtained in section 3. The time of flight spectra have the form shown in Figure 5. There is a contribution to the data from the heavier elements in the sample and sample environment (container, cryostat etc), which must be removed. There is also a small correction for "Final State Effects" (FSE) which is applied to account for the fact that the impulse approximation is not exact at the finite momentum transfer of the measurements.

FSE are subtracted from the hydrogen peak using the method described in Appendix 3. The subtraction of FSE is to some extent cosmetic. A function which is anti-symmetric in momentum space is subtracted. However since the function used to fit the data in is symmetric in momentum space the fitted parameters are virtually unaffected by this step. The quality of the fit is affected however. The FSE subtraction is also useful for a visual assessment of the data. Both these data corrections are made using the command FSESUB

```
eVS> FSESUB
```

Name of file containing time of flight data?

Zh

This is the file name for the corrected data obtained in section 3. The peak you wish to analyse must be the first mass in the file ZH.IN.

input file is zh

```
number of points=    1024
ndmin=    135 ndmax=    182
Name of file containing fit parameters?
```

zh.in

This is the file used for fitting the forward angle data, which was created in section 4d.

```
de1g= 74.00000    de1l= 24.00000
e1= 4897.400
de1g= 74.00000    de1l= 24.00000
ipno=    4
npeaks=    3
    1 am= 1.007900    xs= 0.0000000E+00 wid= 0.0000000E+00
    2 am= 27.00000    xs= 0.3740000    wid= 11.30000
    3 am= 91.00000    xs= 0.6250000    wid= 25.90000
```

NO BACKGROUND SUBTRACTED

BAD PTS NOT EXCLUDED

NO MULTIPLE SCATTERING SUBTRACTED

FSE SUBTRACTED

First and last spectrum number?

135 182

The output file has the name TEMPFSE.DAT.. This contains the time of flight spectra with only the hydrogen peak included. The data is also corrected for deviations from the impulse approximation. The data can be viewed using the command PLOTD.

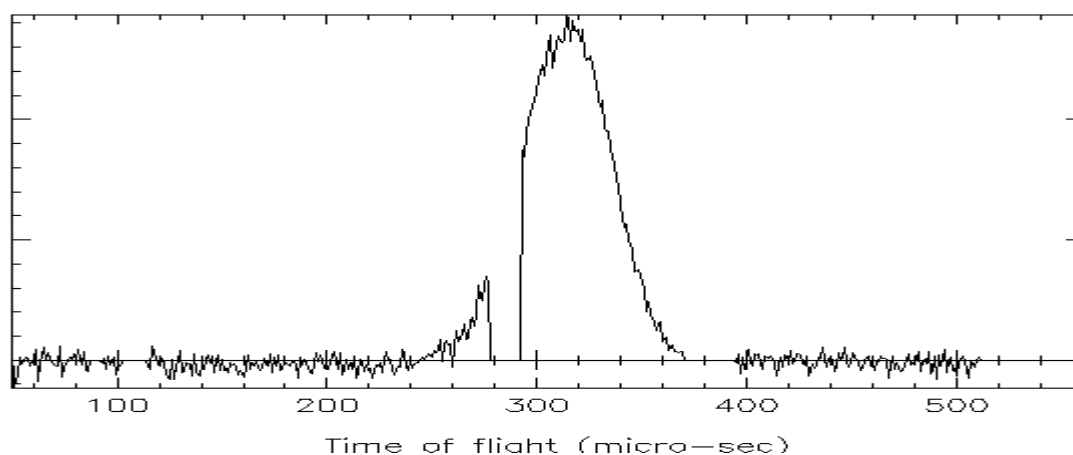


Figure 11. Output of PLOTD on TEMPFSE.DAT for angles 30-40 degrees. The regions which were excluded by PSUBD and FSESUB appear as gaps.

## 5b Creation of file for fitting the hydrogen peak shape.

The next step in the analysis is to create a data file suitable for input to the routine ISOFITU, described in the following section. This is done using the command ISOFILE.

eVS> ISOFILE

Name of file containing time of flight data?

TEMPFSE

input file is zhs

number of points= 512

ndmin= 135 ndmax= 182

Name of file containing fit parameters?

zh.in

de1g= 74.00000 de1l= 24.00000

e1= 4897.400

npeaks= 3

1 am= 1.007900 xs= 0.000000E+00 wid= 0.000000E+00

Note that the first mass in ZHF.IN must be the mass of the atom for which n(p) is to be determined.

2 am= 27.00000 xs= 0.3740000 wid= 11.30000

3 am= 91.00000 xs= 0.6250000 wid= 25.90000

First and last spectrum number?

135 182

.....

ISOFILE normalises the data by

- (i) Transforming the time of flight data to momentum (y) space.
- (ii) Fitting the hydrogen peak area in y space.
- (iii) Dividing the time of flight data by the fitted peak area in momentum space. The latter procedure removes the effects of different detector efficiencies and solid angles.

ISOFILE plots the data and fits in the proton momentum space. It is immediately apparent from these plots if the data analysis procedures and corrections have been successful. In particular since the data is a momentum distribution, it should be symmetric about the origin. To check whether this is so, the program symmetrises the data about the origin and plots the symmetrised (Blue) and unsymmetrised (Black) data (see Figure 11). The sum of fits in the range chosen is also displayed as a red line. These should all coincide within error if the data processing has been successful.

plot data and fit in momentum space

There are 2 options;

1. Plot individual detector
2. Plot sum of consecutive detectors

1

spectrum number? 0 to exit

135

The program allows individual spectra and the sum of spectra to be displayed in momentum space. Figure 12 shows the data from the single spectrum S135.

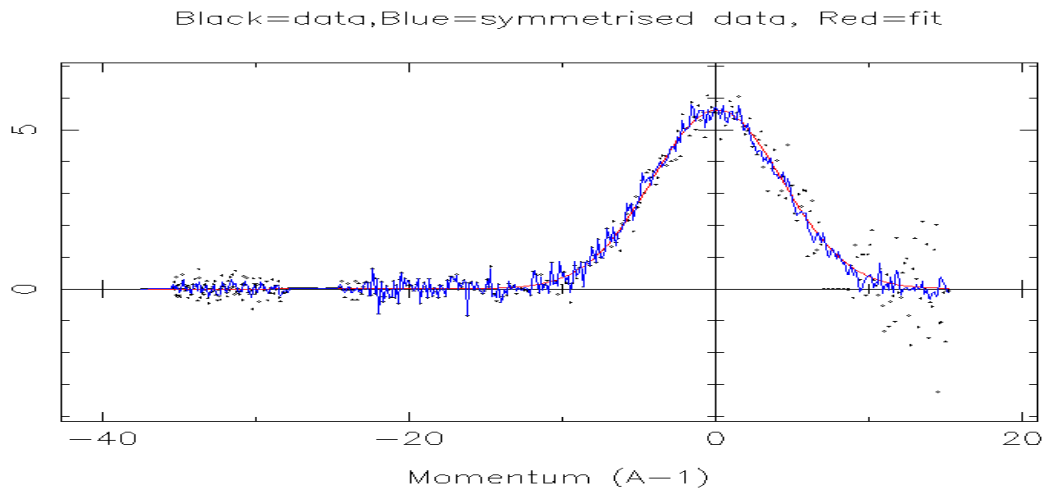


Figure 12. Shows the data in the proton momentum space from detector 135. The black dots are the data. The blue line is the data after symmetrisation about zero. The red line is the fit in momentum space used to normalise the data set. If the procedure has worked properly the red and blue lines should coincide within error.

RETURN

plot data and fit in momentum space

There are 2 options;

1. Plot individual detector
2. Plot sum of consecutive detectors

2

ismin,ismax?

1. 135-142
  2. 143-150
  3. 151-158
  4. 159-166
  5. 167-174
  6. 175-182
  7. 183-190
  8. 191-198
- Type 0 to exit plotting

135 182

Option 2 allows the data to be summed in momentum space. This reduces the statistical errors and gives a more sensitive and more global assessment of the success of the data correction procedures. The list above gives the spectrum number for the 8 banks of detectors at forward scattering angles (see Figure A1.1).

ismin,ismax?

1. 135-142
2. 143-150
3. 151-158
4. 159-166
5. 167-174
6. 175-182
7. 183-190
8. 191-182

0 0

There are 2 options;

1. Plot individual detector
2. Plot sum of consecutive detectors

Type 0 to exit plotting

0

Enter generic file name

zh

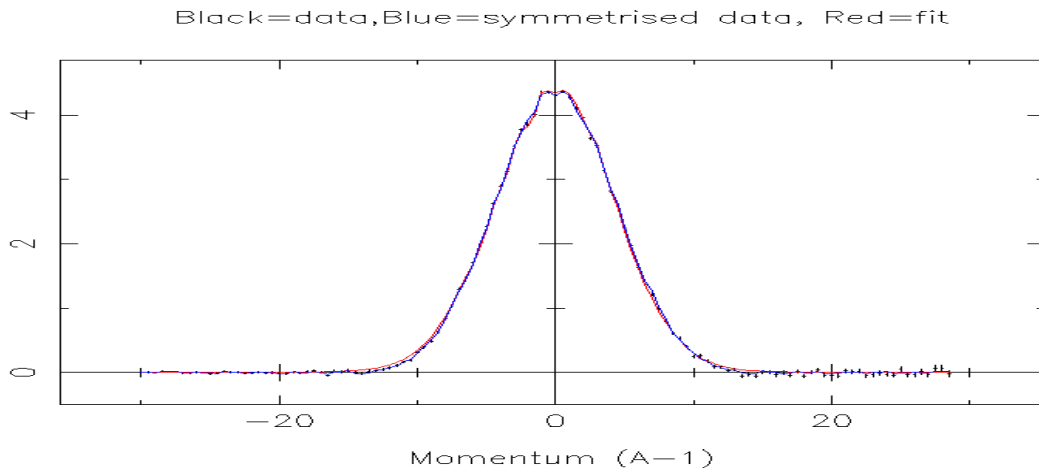


Figure 13 shows sum of detectors 135-182 in the proton momentum space as the black dots. The statistical error bars are also shown. The blue line is the data after symmetrisation about zero. The red line is the sum of fits to individual detectors.

The program produces a file with the name ZHFILE.DAT (XXXXFILE.DAT for generic file name XXXX). This contains the time of flight spectra for every detector included in the input to FSESUB. The data file TEMPFSE.DAT produced by FSESUB should now be deleted to conserve disk space.

```
eVS> delete TEMP*.dat;*
```

### 5c Determination of the proton momentum distribution

The momentum distribution of the protons is obtained using the ISOFITU command. This simultaneously fit all detectors in the data file produced by ISOFILE using the procedure given in Appendices 2- 4. The routine ISOFITU simultaneously fits data from all detectors, to a product of a Gaussian function with a sum of Hermite polynomials. The fitting parameters are the Gaussian width and the coefficients multiplying the Hermite polynomials included. With sufficient coefficients, the fitting function can accurately describe any functional form. However in practice it is rare that more than 5 fitting parameters are required to fit the entire data set.

The first step is to produce a file ZHFIT.IN for input to ISOFITU. This is done using the command CRENPIN (create NP file.IN).

```
eVS> CRENPIN
```

enter generic filename

zh

Mass of atom? (H=1.0079,D=2.015)

1.0079

This sequence of inputs produces a file named ZHFIT.IN of the form

```
zh      File.dat
1.007900
4897.4 24.00 73.6
40.0
zh      fit
410  ! Number of iterations
1.00 1.0 1. Scale factor for xz plane
4.00 1.0 2. Sigma
0.00 0.0 3.          h2*Y00
0.70 1.0 4.          h4*y00
0.00 0.0 5.          h6*Y00
0.00 0.0 6.          h8*y00
0.00 0.0 7.          h10*Y00
0.00 0.0 8.          h12*Y00
0.00 0.0 9.          h14*Y00
0.30 1.0 10.         h3*Y00*sigma/q
0.00 0.0 11.         h4*Y00*sigma**2/q/q
```

If your generic file name is XXXX then the file XXXXFIT.IN will be produced. The different lines in the file are

```
zh      File.dat
```

This is the name of the data file produced by ISOFILE – ZHFILE.DAT. It is the data file which is fitted.

1.007900

This is the atomic mass in amu for which the momentum distribution is to be obtained.. It should be 2.0115 if fitting D.

4897.400000000000 24.0000000000000 73.6000000000000  
40.0000000000000

This is the final energy in meV, the width of the Lorentzian and Gaussian resolution components. 40.0 specifies the range of momentum in output files.

```
zh      fit
```

This specifies the name of the data files output by the fitting routine. These are ZHFITJY.DAT,ZHFITNP.DAT,ZHFITYDAT.DAT.

410

This is the number of iterations of the fitting routine. 410 is adequate in most cases.

1.0 1.0 1. Scale factor for xz plane

This is the scale parameter for fit – this should always be very close to 1.

4.00 1.0 2. Sigma

The standard deviation of the Gaussian in the fitting expansion.

0.00	0.0	3.	$h2*Y00$
0.70	1.0	4.	$h4*y00$
0.00	0.0	5.	$h6*Y00$
0.00	0.0	6.	$h8*y00$
0.00	0.0	7.	$h10*Y00$
0.00	0.0	8.	$h12*Y00$
0.00	0.0	9.	$h14*Y00$

Coefficients of Hermite polynomials; The first number is the starting values of the fitting parameter. If the second number is 1 the parameter is fitted. If it is zero it is fixed.

0.30	1.0	10.	$h3*Y00*\sigma/q$
------	-----	-----	-------------------

Fits any asymmetries in the data set. If the assumptions of the data analysis are correct such asymmetries should be very small. If results depend significantly upon whether this parameter is included in the fit, the data is suspect.

0.00	0.0	11.	$h4*Y00*\sigma^2/q/q$
------	-----	-----	-----------------------

Corrects for second order final state effects not included in the FSE corrections of section 5a. This parameter is usually set to zero and not fitted as second order corrections are unobservable at current statistical accuracies.

The default is to fit only the scale factor, the Gaussian width and the coefficient to H4. However your data may contain more structure, which need more coefficients to fit. The coefficients which should be included are determined automatically using the command CHOOSEPAR.

```
eVS> choosepar
enter input file name
zhfit
```

CHOOSEPAR calculates the likelihood of the various combinations of the coefficients of H2-H14. A better fit is always obtained by including more parameters, but eventually one ends up fitting noise in the data. CHOOSEPAR uses a Bayesian method to limit the number of fitting parameters in a systematic way. It calculates the likelihood of the fit using the “Occams razor” method described in Reference [7]. CHOOSEPAR takes about 10 minutes to run and produces a file CHOOSEPAR.OUT containing the likelihoods of all combinations of parameters.

A file for fitting with the parameters determined as significant by CHOOSEPAR can be created using the command SORT\_CHOUSEPAR.

```
eVS> sort_choosepar
```

```
Input file was ZHFIT
```

	log lik	H2	H4	H6	H8	H10	H12
2	-6418.521	0	1	0	0	0	0
4	-6419.234	0	0	1	0	0	0
8	-6420.410	0	0	0	1	0	0

.....

The routine prints out the various combinations of parameters in order of their likelihood. Only the first few lines of the output are shown. The first line gives the optimum combination of fitting parameters. 1 means the parameter should be included, 0 that it should not. It then prompts for the generic filename and mass and produces a new ZHFIT.IN. This includes only the coefficients giving the maximum likelihood in the fit.

```
Generic file name?
```

```
zh
```

```
Mass of atom? (H=1.0079,D=2.015)
```

```
1.0079
```

The final stage of the analysis is to determine the function  $J(y)$  and the corresponding momentum distribution by simultaneously fitting the same parameters to data in all detectors.

```
eVS> ISOFITU
```

```
enter input file name
```

```
ZHFIT
```

This is the file produced by SORT\_CHOUSEPAR.

When the program has found the optimum values of the fit parameters it gives a prompt;

```
VALUE OF RED CHISQ= 1.06207331146182
```

```
Plot fitted J(y) with errors
```

```
Graphics device/type (? to see list, default /NULL):
```

(A .gif file of this plot can be obtained by entering /GIF, POSTSCRIPT file by entering /PS.)



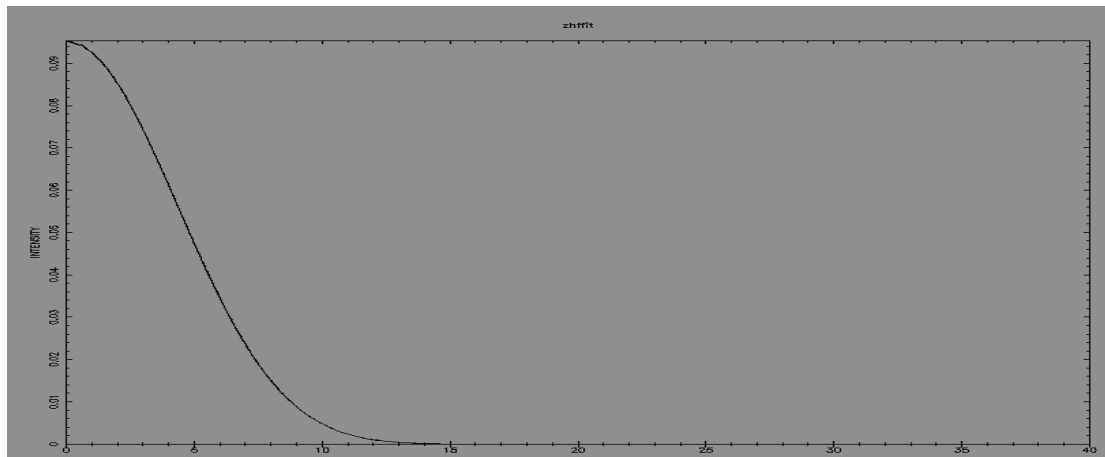


Figure 14. Compton profile  $J(y)$  produced by ISOFITU.

A plot of the fitted  $J(y)$  with errors will appear on the screen as shown in Figure 14.

The program also writes an Ascii file of the plot to ZHFITJY.DAT in the format  
 $y$ value,  $J(y)$  error in  $J(y)$ .

Next the program gives the prompt

Type <RETURN> for next page:

Plot is of  $p^2 n(p)$

Graphics device/type (? to see list, default /NULL): /XW

A plot of  $p^2 n(p)$  will appear, where  $n(p)$  is the momentum distribution.

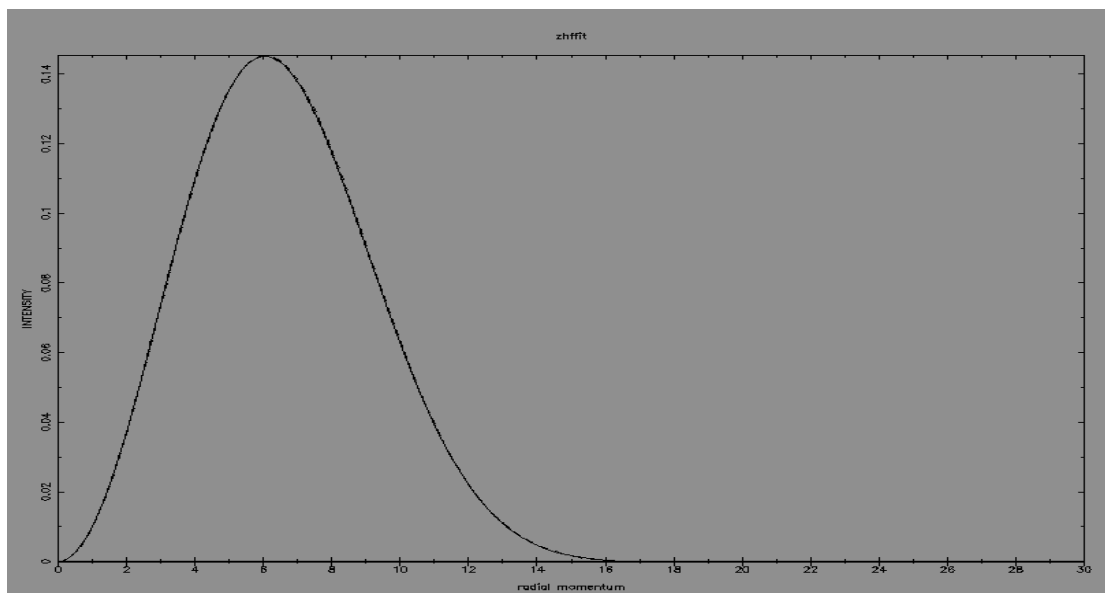


Figure 15. Plot of  $p^2 n(p)$  produced by ISOFIT. The errors are also plotted but are too small to be observed.

The program also produces an ASCII file ZHFITNP.DAT. in the form

$p$ ,  $p^2 n(p)$ , error in  $p^2 n(p)$ .

The next prompt is

PLOT Y1 AND Y2 (1)

PLOT RESIDUAL(2)

FOR NO PLOT (0)

This gives a plot of the data and fits after all detectors have been added in y space

A response of **1** will give a plot of the experimental  $J(y)$  convoluted with the resolution function, together with the fit.

A response of **2** will give a plot of the difference between experiment and fit

A response of **0** will give no plot and the program will move to the next stage.

The program also produces an ASCII file ZHFITYDAT.DAT which contains the data and fit in yspace. This is of the form

yvalue, data, fit, statistical error in data point.

The next prompt is

Plotfile? (1=yes, 0=no)

**1**

PLOTFILE BEING CREATED.

This produces a file TEMPFILE.DAT which contains the data and fit to each individual detector in time of flight. This is useful to assess the quality of the fits and to see if particular detectors are giving poor fits. The TEMPFILE.DAT file can be examined using the command

**\$ PLOTDETS**

A list of values of the reduced chi-square for each detector is given.

1	48	512
1	135	Reduced chi-square of fit= 0.8686708
2	136	Reduced chi-square of fit= 0.9556400
3	137	Reduced chi-square of fit= 0.8972189
4	138	Reduced chi-square of fit= 0.8466188
5	139	Reduced chi-square of fit= 0.9704176
6	140	Reduced chi-square of fit= 1.013893

The individual fits can also be plotted. The program gives the prompt

DETECTOR NUMBER? (0 TO END)

**40**

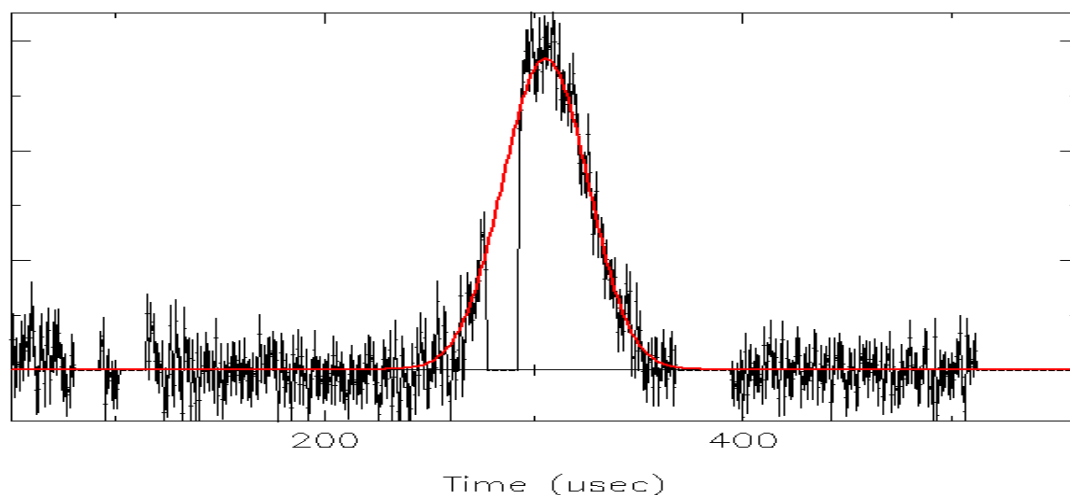


Figure 16. Shows the data and fit in detector number 40 of the 48 detectors included. The data is plotted as black points with error bars. The fit is the solid red line. The points which are excluded from the fit by the routines FSESUB and PSUBD appear as gaps in the data.

After the fits to individual detectors have been examined all data files with names beginning with TEMP should be deleted to conserve disk space. This can be done by the command.

```
>eVS delete temp*.dat;*
```

If necessary all such deleted files can be recreated very quickly by rerunning the relevant programs.

This completes the analysis procedure. The following Appendices contain more details of the VESUVIO instrument, the correction procedures used and the data analysis procedures.

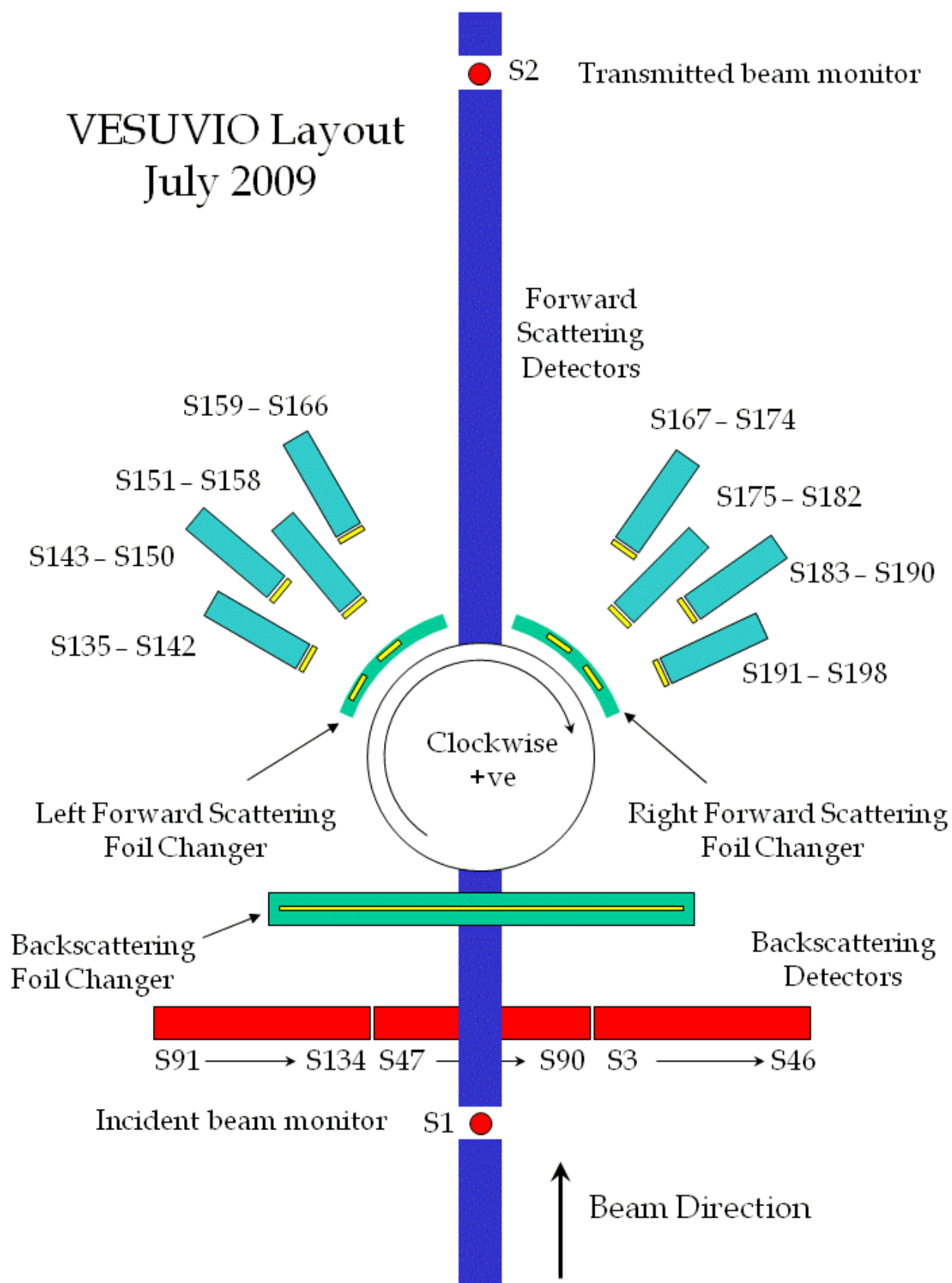


Figure A1.1 shows a schematic diagram of the VESUVIO spectrometer.

## Appendix 1 The VESUVIO Spectrometer

The VESUVIO spectrometer is illustrated in Figure A1.1. VESUVIO at present has 198 detectors.

1. Spectrum 1 (S1) is the incident beam monitor.
2. S2 is the transmitted beam monitor.
3. S3-S134 are back scattering detectors. These lie in the angular range 133-165 degrees.
4. S135-198 are forward scattering detectors. These lie in the angular range 30-70 degrees.

VESUVIO measures the momentum distribution  $n(\mathbf{p})$  of atoms by "neutron Compton scattering" (NCS) [8]. Measurements of  $n(\mathbf{p})$  provide a unique window on the quantum behaviour of atomic nuclei in condensed matter systems. NCS is of particular value for studying the large number of important systems containing protons - for example, water, biological molecules, hydrogen storage materials, proton conductors, metal hydrides etc. Examples of recent measurements in various systems are given in references [9, 10, 11, 12, 13, 14, 15].

Interpretation of VESUVIO data relies upon the fact that for scattering with sufficiently high momentum and energy transfers, the impulse approximation (IA) is accurate [22]. The IA implies that the neutron scatters from a single atom, with conservation of the total kinetic energy and momentum of the neutron plus atom. Hence the measured energy and momentum change of the neutron is related in a simple way to the momentum distribution of the atoms. Details of the technique and the data analysis used are given in references [8] and [16] and are also summarised in Appendix 2.

Measurements of proton momentum distributions require much higher energy and wave vector transfers than those accessed by other neutron scattering techniques. Typically the energy transfer is  $1 < \omega < 50$  eV and the wave vector transfer  $40 < q < 150$  Å<sup>-1</sup>. NCS is possible only at pulsed neutron sources, which allow the use of time of flight techniques [17] for accurate inelastic neutron scattering measurements at eV energies.

Time of flight techniques require that the energy of either the incident or scattered neutron is well defined. On VESUVIO this is accomplished by use of the neutron absorption resonance which occurs in gold at 4.9 eV. This defines the final energy of the neutron as  $4.9 \pm \sim 0.14$  eV. The data produced by VESUVIO is obtained by taking differences between spectra produced with gold foils in different positions. The foil changers are also illustrated in Figure A1.1.

To illustrate the differencing method we consider data at forward scattering. The technique at back scattering is discussed in reference [1]. The VESUVIO detectors at forward scattering angles are Yttrium Aluminium Perovskite doped (YAP)  $\gamma$ -ray detectors [18,19,20]. Although these do not detect neutrons directly, they are made to function both as neutron detectors and energy selectors by placing a gold foil on the YAP detector surface [21]. Gold contains a sharp resonance absorption line for neutrons, centred at 4.9 eV. When the foil absorbs neutrons in the 4.9 eV resonance it emits a  $\gamma$ -ray cascade which can be detected, thereby determining both the time of flight and the final energy of the neutron.

The blue line in Figure A1.2 shows time of flight data obtained in a single YAP detector at a scattering angle of  $66^\circ$ , from a sample composed of sheets of lead and polythene ( $\text{CH}_2$ ). The peak at  $\sim 380 \mu\text{sec}$  is due to scattering from lead and carbon, that at  $\sim 170 \mu\text{sec}$  from hydrogen. It can be seen that the  $\gamma$ -ray signal from the foil sits on a large background. This seems to be mainly due to  $\gamma$ -ray rays produced when neutrons scattered by the sample are absorbed by the surrounding boron shielding. There are also a series of lines for  $t < 200 \mu\text{sec}$  due to absorption in antimony impurities in the lead sample.

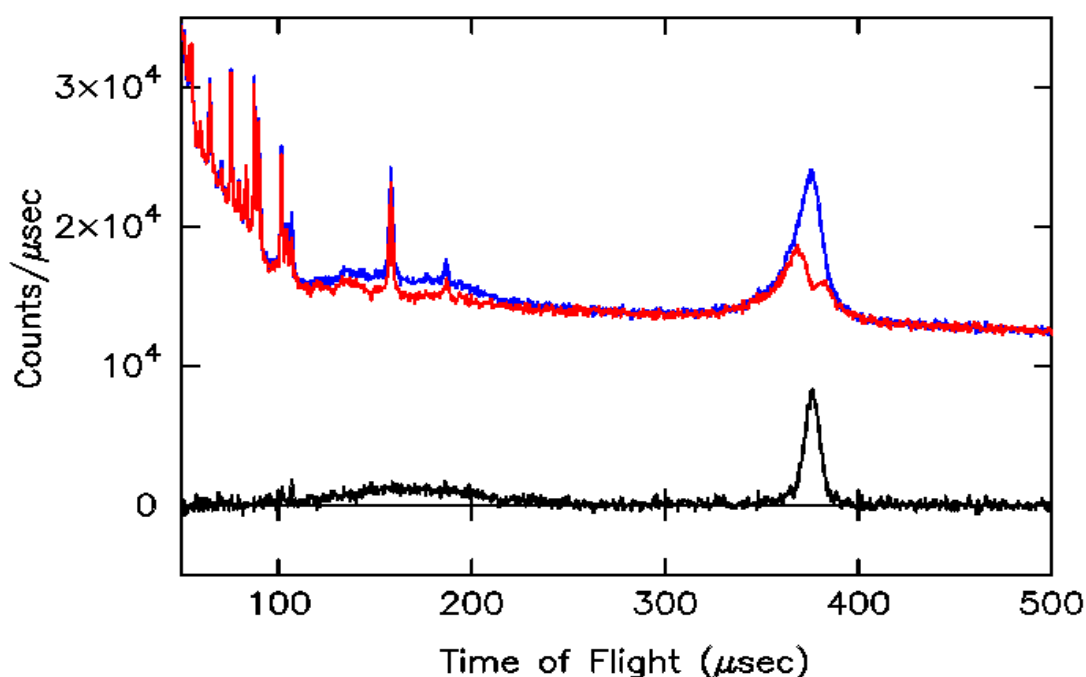


Figure A1.2 Data taken from a sample composed of sheets of polythene ( $\text{CH}_2$ ) and lead by a single YAP detector at an angle of  $67^\circ$ . The "foil out" data is in blue. The "foil in" data is in red. The difference "foil out"-"foil in" is shown as the black line. The scattering angle was  $66^\circ$ . The total count time was 5600 micro-amp-hours. The sample scattered 15% of the incident beam.

By use of a secondary foil which can be placed in two positions the energy resolution can be much improved and the background almost eliminated. This "foil cycling" method [21] is illustrated in Figure A1.3.

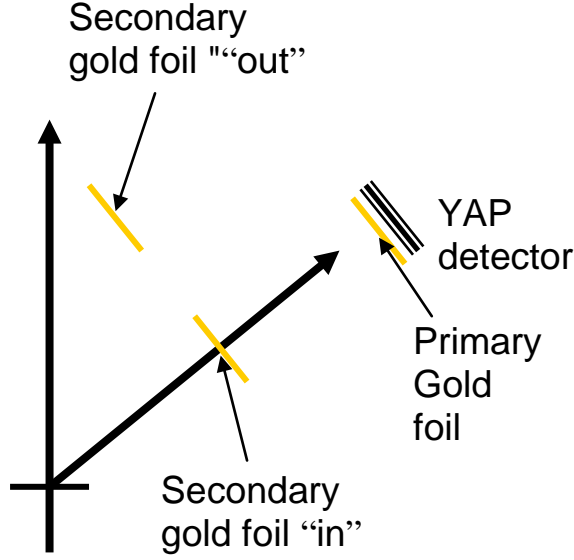


Figure A1.3. Illustrates the foil cycling method used on VESUVIO. There is a primary gold foil fixed on the surface of the YAP detector and a moveable secondary gold foil of identical thickness. The secondary foils are "cycled" that is moved many times between the two positions within a data collection period. The cycling removes drifts in detector efficiency with time, due for example to ambient temperature changes.

In the "foil out" measurement (shown as the blue line in Figure A1.2), the scattered neutrons do not have to pass through the secondary foil to reach the primary foil. The "foil out" counts are

$$C_{out}(E_0, E_1, \theta_d) = C(E_0, E_1, \theta_d)A(E_1)\eta + B_{out}(E_0, E_1, \theta_d) \quad (A1.1)$$

$E_0$  is the energy of the neutron incident on the sample,  $E_1$  is the energy of the scattered neutron absorbed by the foil and  $\theta_d$  is the scattering angle.  $C(E_0, E_1, \theta_d)$  is the rate at which scattered neutrons are incident on the primary foil.  $C(E_0, E_1, \theta_d)$  depends on the incident neutron spectrum and the scattering properties of the sample and can be determined from the data.  $A(E_1)$  is the probability that a neutron of energy  $E_1$  is absorbed in the primary foil.  $\eta$  is the probability that the consequent  $\gamma$ -ray cascade is registered by the YAP detector.  $B_{out}$  is the  $\gamma$  background with the secondary foil "out".

In the "foil in" measurement the secondary foil is moved so that neutrons have to pass through it to reach the primary foil. Currently the secondary foil has the same thickness as the primary foil. Hence the counts from the primary foil are reduced by the factor  $1 - A(E_1)$ .

$$C_{in}(E_0, E_1, \theta) = C(E_0, E_1, \theta)[1 - A(E_1)]A(E_1)\eta + B_{in}(E_0, E_1, \theta) \quad (A1.2)$$

A typical "foil in" spectrum is shown in Figure A1.2 as the red line. The VESUVIO "raw" data is the difference between these two counts.

$$(C_{out} - C_{in}) = C\eta A^2 + (B_{out} - B_{in}) \quad (A1.3)$$

This is illustrated as the black line in Figure A1.2. Clearly most of the  $\gamma$ -ray background is removed by this differencing technique. The residual background ( $B_{out} - B_{in}$ ) is calculated by the BCORR routine of section A10.3, using the method described in reference [2].

It can be seen from eqs (A1.1) and (A1.3) that the (almost) Lorentzian energy dependence of  $A(E_1)$  in the "foil out" measurement becomes a Lorentzian-squared dependence in the difference measurement. Thus not only does the foil cycling method remove most of the gamma background but it also greatly improves the resolution function. This is illustrated in Figure A1.4, which shows lead data collected with the old  $^6\text{Li}$  doped neutron detectors [1] and the new YAP system as a function of energy transfer,  $(E_0 - E_1)$ . The energy resolution function with the  $^6\text{Li}$  detectors is almost Lorentzian with a FWHM of  $\sim 0.35$  eV. The energy resolution with the YAP detectors is almost Gaussian with a FWHM of  $\sim 0.2$  eV [3].

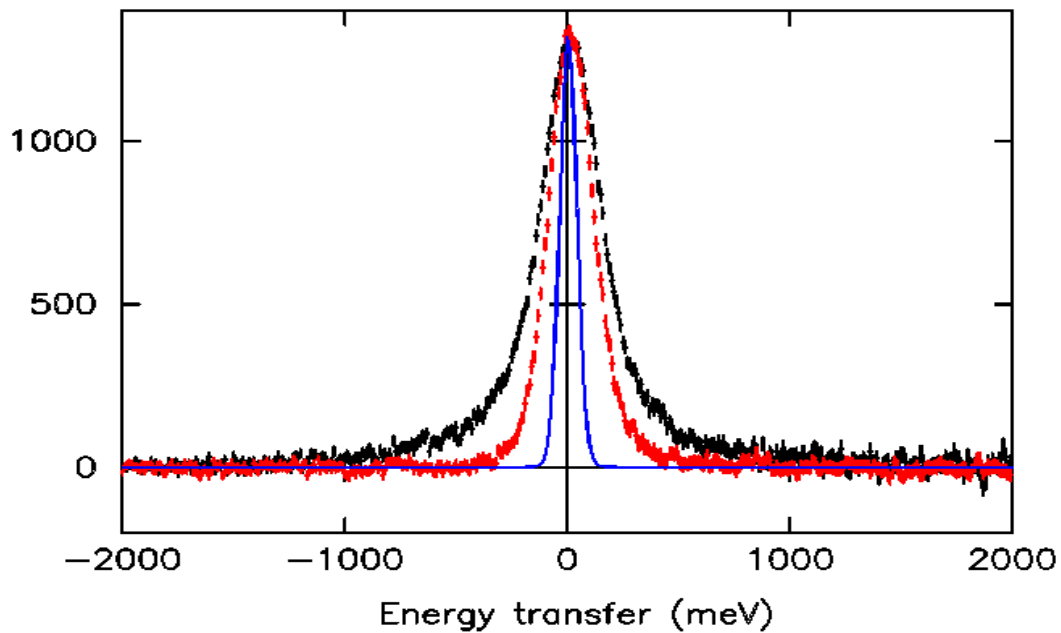


Figure A1.4. Shows VESUVIO data collected on a Pb sample with the old  $^6\text{Li}$  doped neutron detectors (black) and the new YAP detector system (red) as a function of energy transfer  $(E_0 - E_1)$ . The intrinsic width due to the lead momentum distribution is shown as the blue line.

At present on VESUVIO the back-scattering detectors use the older double difference technique [4]. This gives two measurements with different resolutions and count-rates - the single difference and the double difference spectra. The double difference data which is usually used gives similar energy resolution to that obtained at forward scattering.



## Appendix 2. Theory of the time of flight data analysis.

### A2a Count rates on inverse geometry spectrometers

We consider a system of  $N$  identical atoms, scattering neutrons into a detector subtending solid angle  $d\Omega$ , at scattering angle  $\theta$ . It follows from the definition [22] of the partial differential scattering cross-section  $d^2\sigma / d\Omega dE$ , that the number of neutrons with incident energies in the range  $E_0$  to  $E_0 + dE_0$ , detected with final energies between  $E_1$  and  $E_1 + dE_1$  is

$$C_D(E_0, E_1) dE_0 dE_1 = I(E_0) D(E_1) \frac{d^2\sigma(E_0, E_1, \theta)}{d\Omega dE_1} d\Omega dE_0 dE_1 \quad (\text{A2.1})$$

where  $I(E_0)dE_0$  is the number of incident neutrons/unit area with energies between  $E_0$  and  $E_0 + dE_0$  and  $D(E_1)$  is the probability that a neutron of energy  $E_1$  is detected. It follows from standard theory [22] that for isotropic scattering,

$$\frac{d^2\sigma(E_0, E_1, \theta)}{d\Omega dE_1} = |b|^2 \sqrt{\frac{E_1}{E_0}} S(q, \omega) \quad (\text{A2.2})$$

where  $S(q, \omega)$  is the dynamic structure factor, and  $b$  is the nuclear scattering length. The energy transfer in the measurements is

$$\omega = E_0 - E_1 \quad (\text{A2.3})$$

and the momentum transfer

$$q = \sqrt{2m} (E_0 + E_1 - 2\sqrt{E_0 E_1} \cos \theta)^{1/2} \quad (\text{A2.4})$$

The velocity of the scattered neutron is

$$v_1 = \sqrt{2E_1 / m} \quad (\text{A2.5})$$

with a similar expression for the velocity  $v_0$  of the incident neutron, where  $m$  is the neutron mass. The neutron time of flight  $t$  is thus

$$t = \frac{L_0}{v_0} + \frac{L_1}{v_1} \quad (\text{A2.6})$$

where  $L_0$  is the incident flight path and  $L_1$  is the final flight path. Equations (A2.5) and (A2.6) can be used to define  $E_0$  in terms of  $E_1$  and  $t$ .

$$E_0(E_1, t) = \frac{m}{2} \left( \frac{L_0 v_1}{v_1 t - L_1} \right)^2 \quad (\text{A2.7})$$

The total number of neutrons detected in a time channel between  $t$  and  $t + dt$  can be expressed as

$$C(t)dt = \left[ \int C_D[E_0(t, E_1), E_1] \frac{dE_0(t, E_1)}{dt} dE_1 \right] dt \quad (\text{A2.8})$$

It follows from (A2.5) and (A2.7) that

$$\frac{dE_0}{dt} = \left( -\frac{2^{3/2}}{L_0 m^{1/2}} \right) E_0^{3/2} \quad (\text{A2.9})$$

On an ideal inverse geometry instrument,  $L_0, L_1, \theta$  are precisely known and only neutrons of a precisely defined energy  $E_R$  are detected - that is

$$D(E_1) = D(E_R) \delta(E_1 - E_R) \quad (\text{A2.10})$$

It then follows from (A2.1) and (A2.8) – (A2.10) that

$$C(t) = 2 \left( \frac{2}{m} \right)^{1/2} \frac{E_0^{3/2}}{L_0} I(E_0) D(E_R) N \frac{d^2 \sigma}{d\Omega dE_1} d\Omega \quad (\text{A2.11})$$

where  $E_0(E_R, t)$  is defined via (A2.7). Equation (A2.11) is the standard expression for the count rate in an inverse geometry time of flight spectrometer [17].

### A2b. The Impulse Approximation

The VESUVIO spectrometer is mainly used to determine atomic momentum distributions in condensed matter systems, by “Deep Inelastic Neutron Scattering” (DINS), also known as Neutron Compton Scattering (NCS) [8]. DINS relies upon the fact that at sufficiently high momentum transfer, the Impulse Approximation (IA) can be used to interpret neutron data [22]. The validity of the IA in neutron scattering has been discussed by many authors [23,24,25,26] and at the energy and momentum transfers attained on VESUVIO is accurate to within ~5% in hydrogenous samples [27]. A basic assumption of the IA is that for neutron wavelengths much less than the inter-atomic spacing, atoms scatter incoherently. Thus if atoms of different mass  $M$  are present in the sample, it follows from (A2.11) that the count rate is

$$C(t) = 2 \left( \frac{2}{m} \right)^{1/2} \frac{E_0^{3/2}}{L_0} I(E_0) D(E_R) \sum_M N_M \frac{d^2 \sigma_M}{d\Omega dE_1} d\Omega \quad (\text{A2.12})$$

where  $N_M$  is the number of atoms of mass  $M$  and  $d^2 \sigma_M / d\Omega dE_1$  is the partial differential cross-section for mass  $M$ . The IA effectively treats the scattering as single atom “billiard ball” scattering with conservation of momentum and kinetic energy of the neutron + target atom. The formal expression of this is that the dynamic structure factor for atoms of mass  $M$  is [22]

$$S_M(\mathbf{q}, \omega) = \int n_M(\mathbf{p}) \delta \left( \omega + \frac{p^2}{2M} - \frac{(\mathbf{p} + \mathbf{q})^2}{2M} \right) d\mathbf{p} \quad (\text{A2.13})$$

where  $n_M(\mathbf{p})$  is the atomic momentum distribution for mass  $M$ .

It follows from (A2.13) that

$$S_M(\mathbf{q}, \omega) = \frac{M}{q} J_M(y_M, \hat{\mathbf{q}}) \quad (\text{A2.14})$$

where

$$y_M = \frac{M}{q} \left( \omega - \frac{q^2}{2M} \right). \quad (\text{A2.15})$$

and

$$J_M(y_M, \hat{\mathbf{q}}) = \int n_M(\mathbf{p}) \delta(y_M - \mathbf{p} \cdot \hat{\mathbf{q}}) d\mathbf{p} \quad (\text{A2.16})$$

where  $\hat{\mathbf{q}} = \mathbf{q}/q$ . The “neutron Compton profile”  $J_M(y_M, \hat{\mathbf{q}})$  is the probability distribution of the momentum component of mass  $M$  along the direction of  $\hat{\mathbf{q}}$  and is analogous to the “Compton profile”, measured in Compton scattering of photons from electrons. In isotropic samples such as liquids and powders, all directions are equivalent and the dependence upon  $\hat{\mathbf{q}}$  can be ignored.

In the latter case it follows from equation (A2.14) and (A2.2) that,

$$\frac{d^2 \sigma_M}{d\Omega dE_1} = b_M^2 \sqrt{\frac{E_1}{E_0}} \frac{M}{q} J_M(y_M) \quad (\text{A2.17})$$

Combining equations (A2.12) and (A2.17)

$$C(t) = \frac{E_0 I(E_0)}{q} \sum_M A_M M J_M(y_M) \quad (\text{A2.18})$$

where

$$A_M = \frac{2}{L_0} D(E_R) \sqrt{\frac{2E_R}{m}} \Delta\Omega N_M b_M^2 \quad (\text{A2.19})$$

is proportional to number of atoms of mass  $M$  multiplied by the scattering cross-section.

## 2c. Fitting Expression

In the derivation of equation (A2.18) it is assumed that the “instrument parameters”  $L_0, L_1, \theta$  and  $E_1$  are known exactly. In reality these quantities can be determined only according to some probability distribution  $P(L_0, L_1, \theta, E_1)$ , which determines the instrument resolution.

The measured count rate  $C_m(t)$  is an average over the possible values of these parameters, weighted by their probability of occurrence

$$C_m(t) = \int C(t) P(L_0, L_1, \theta, E_1) dL_0 dL_1 d\theta dE_1 \quad (\text{A2.20})$$

Thus the exact incorporation of the instrument resolution function would entail the evaluation of this four dimensional integral for each data point, in addition to the convolution in  $t$ , required to incorporate the uncertainty in the measurement of time of flight. To reduce data processing times, the approximation is made in the data analysis that the resolution can be incorporated as a single convolution in  $t$  space, with a different resolution function  $R_M(t)$  for each mass. Thus (A2.18) is modified to

$$C_m(t) = \left[ \frac{E_0 I(E_0)}{q} \right] \sum_M A_M M J_M(y_M) \otimes R_M(t) \quad (\text{A2.21})$$

A second approximation of the data analysis is that  $J_M(y_M)$  is assumed to have a normalised Gaussian form

$$J_M(y_M) = \frac{1}{\sqrt{2\pi w_M^2}} \exp\left(-\frac{y_M^2}{2w_M^2}\right) \quad (\text{A2.22})$$

Equations (A2.21-22) define the fitting expression in the routines TFIT, used to fit data in time of flight. The parameters fitted are  $A_M$ , the amplitudes of scattering from each peak and  $w_M$ , which defines the Gaussian width of each peak in momentum ( $y_M$ ) space

### Appendix 3. Corrections to the Impulse Approximation

The method of Sears [23] is used to correct data for deviations from the impulse approximation, due to the non-infinite momentum transfer  $q$  and energy transfer  $\omega$  in the measurement. Sears showed that the effects of finite  $q$  and  $\omega$  can be accounted for by writing  $J(y)$  as,

$$J(y) = J_{IA}(y) - \frac{M \langle \nabla^2 V \rangle}{36 \hbar^2 q} \frac{d^3 J_{IA}(y)}{dy^3} + \frac{M^2 \langle F^2 \rangle}{72 \hbar^4 q^2} \frac{d^4 J_{IA}(y)}{dy^4} + \dots \quad (\text{A3.1})$$

where  $J_{IA}(y)$  is the impulse approximation result.  $\langle \nabla^2 V \rangle$  is the mean value of the Laplacian of the potential energy of the atom and  $F$  is the force on the atom. If  $q$  is sufficiently large, all terms other than  $J_{IA}(y)$  can be neglected. In the VESUVIO data analysis routines only the first and second terms on the right in eq. (A3.1) are included in the analysis. The third term is negligible at the current level of statistical accuracy. The value of  $\langle \nabla^2 V \rangle$  is fitted by the routine FSESUB and the second term on the left in eq (A3.1) is subtracted from the data.

The validity of this method has been tested on zirconium hydride [27] beryllium metal and zirconium deuteride. [28]. It has been shown that within experimental error the corrections to

the impulse approximation applied on VESUVIO are consistent with equation (A3.1) in these materials.

#### Appendix 4. Theory of Fitting to obtain $n(\mathbf{p})$ for a single mass

As discussed in Appendix 2, VESUVIO measures not the momentum distribution  $n(\mathbf{p})$ , but the “longitudinal momentum distribution”  $J(y, \hat{\mathbf{q}})$ , where  $\hat{\mathbf{q}}$  is the unit vector along  $\mathbf{q}$ . The two quantities are related via the expression (A2.16)

$$J(y, \hat{\mathbf{q}}) = \int n(\vec{p}) \delta(y - \mathbf{p} \cdot \hat{\mathbf{q}}) d\mathbf{p} \quad (\text{A4.1})$$

where

$$y = \frac{M}{q} \left( \omega - \frac{q^2}{2M} \right). \quad (\text{A4.2})$$

$J(y, \hat{\mathbf{q}})$  is the “Radon transform” of  $n(\mathbf{p})$ . In order to determine  $n(\mathbf{p})$  from  $J(y, \hat{\mathbf{q}})$ , this transform must be inverted. The method used to invert the transform relies upon the following mathematical result [29]. If  $J(y)$  is expressed in the form

$$J(y, \hat{\mathbf{q}}) = \frac{\exp(-y^2 / 2\sigma^2)}{\pi^{1/2}} \sum_{n,l,m} a_{n,l,m} H_{2n+1}(y) Y_{l,m}(\hat{\mathbf{q}}) \quad (\text{A4.3})$$

Then

$$n(\mathbf{p}) = \frac{\exp(-p^2 / 2\sigma^2)}{\pi^{3/2}} \sum_{n,l,m} 2^{2n+l} n! (-1)^n a_{n,l,m} L_n^{l+1/2}(p^2) Y_{l,m}(\hat{\mathbf{p}}) \quad (\text{A4.4})$$

In eqn (A4.3)  $H_{2n+1}$  is a Hermite polynomial,  $Y_{l,m}(\hat{\mathbf{q}})$  is a Legendre polynomial. In eqn A4.4

$L_n^{l+1/2}(p^2)$  is a Laguerre polynomial. Since Hermite polynomials and Legendre polynomials form a complete set, with enough coefficients the expansion (A4.3) can be used to accurately describe any three dimensional function. With isotropic data, where there is no dependence on the direction  $\hat{\mathbf{q}}$ , only the terms  $l, m = 0$  contribute and these equations reduce to;

$$J(y) = \frac{\exp(-y^2 / 2\sigma^2)}{\pi^{1/2}} \sum_n a_n H_{2n+1}(y) \quad (\text{A4.5})$$

$$n(p) = \frac{\exp(-p^2 / 2\sigma^2)}{\pi^{3/2}} \sum_n 2^{2n} n! (-1)^n a_n L_n^{1/2}(p^2) \quad (\text{A4.6})$$

The program ISOFITU fits the time of flight spectra to eq (A2.21) with  $J(y)$  given by eq (A4.5). The fitting parameters are  $\sigma$  and the coefficients  $a_n$ . The momentum distribution

$n(p)$  is calculated by inserting these fitted coefficients in eq (A4.6). The errors in  $n(p)$  are calculated from the statistical errors on the fitted parameters as described in refs [8, 30]. The steps required to run the programs are given in section 5.

## Appendix 5. Multiple Scattering Corrections

Multiple scattering (MS) occurs because there is a finite probability that the neutron will scatter more than once in the sample before being detected. MS must be subtracted from the time of flight spectra before accurate results for momentum distributions can be obtained. The multiple scattering is calculated using the procedure described in reference [6]. This is done in the routine MSSUB described in section A10.5.

In practice probably the most troublesome component of multiple scattering is due to the neutron scattering twice from heavy atoms in the sample. Such scattering gives a spurious peak to the left of the heavy atom contribution in the time of flight spectrum. Multiple scattering in which protons are involved gives a smooth background under the hydrogen peak. This is illustrated in Figure A5.1 which shows the single and multiple scattering from the  $\text{ZrH}_2$  sample used throughout as an example. The simulation was for a sample which scatters 9.2% of the incident neutrons. Spectra 151-158 (42-53 degrees) were summed.

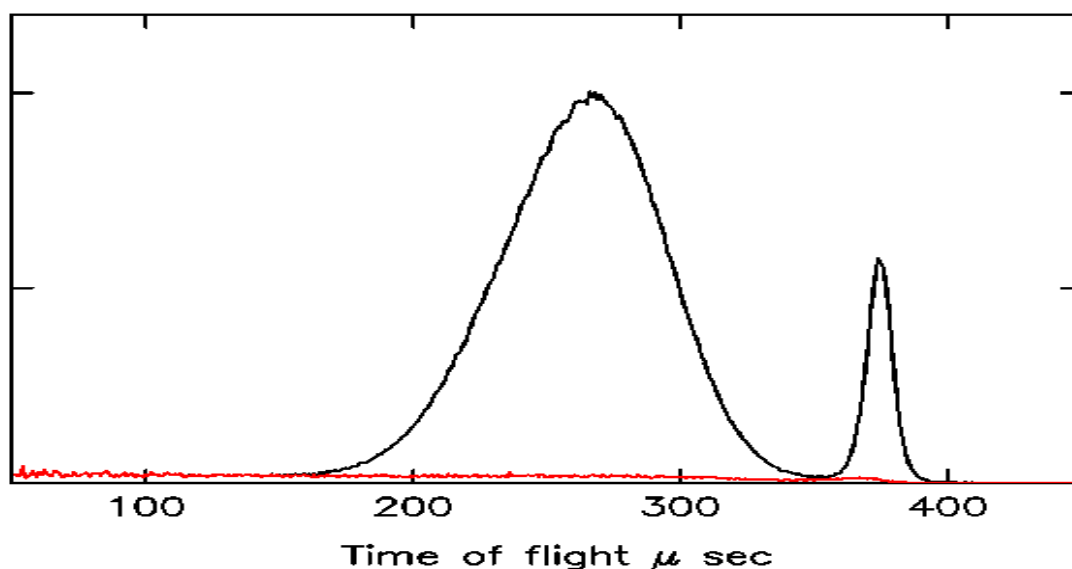


Figure A5.1. Shows calculated total (black line) and multiple scattering (red line) for a  $\text{ZrH}_2$  sample, scattering 9.2% of the incident neutrons. Multiple scattering involving Zr-H, Al-H or H-H gives a broad background under the single scattering from H. The Al-Zr, Al-Al or Zr-Zr multiple scattering gives a secondary peak to the left of the single scattering contribution from Al+La which is centred at  $\sim 370 \mu\text{sec}$ . **Redo for sample used**

The ratio of the intensities of multiple/single scattering is determined by the sample thickness. However the shape of the multiple scattering contribution is quite insensitive to the sample thickness. This is illustrated in figure A5.2 which shows the multiple scattering calculated for

ZrH<sub>2</sub> samples with different thicknesses. The attenuation  $A$  in the sample ( $A=1-\text{transmission}$ ) is given for each case. The multiple scattering is divided by the sample thickness input to the program. It can be seen that the shape of the multiple scattering is independent of sample thickness to a good approximation.

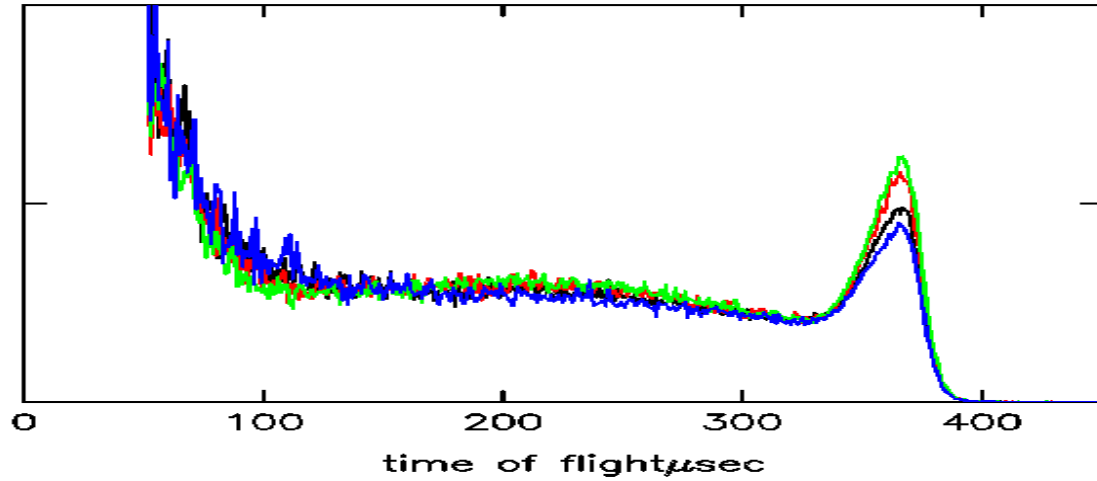


Fig A5.2a. Shows the calculated multiple scattering summed over back scattering spectra 91-134 for ZrH<sub>2</sub> in an Al container. Blue attenuation  $A=0.048$ . Black  $A=0.092$ , Red  $A=0.179$ , Green  $A=0.256$ . The calculated MS is divided by the sample thickness in each case.

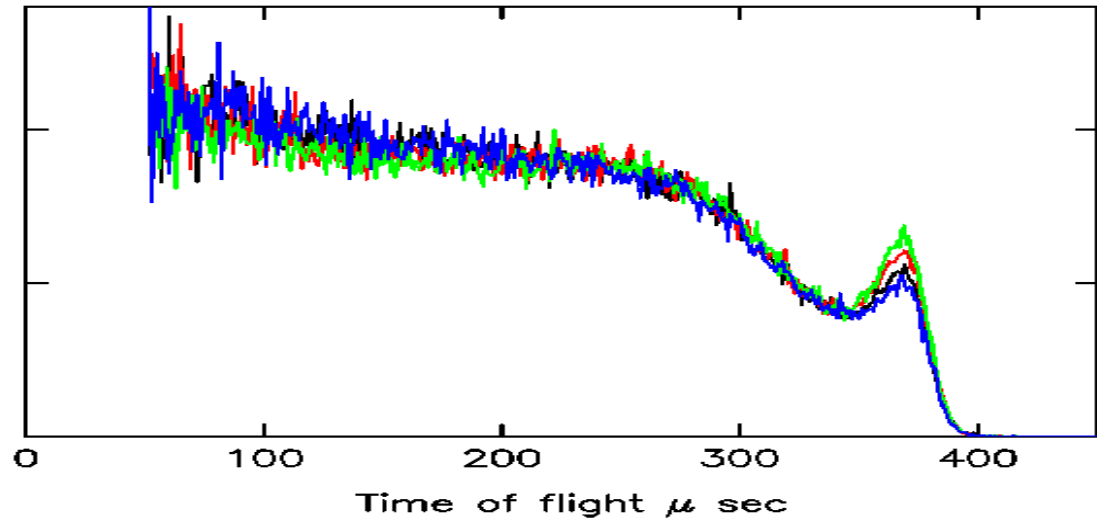


Figure A5.2b. Shows the calculated multiple scattering summed over spectra 135-182 at forward angles. Blue  $A=0.048$ . Black  $A=0.092$ , Red  $A=0.179$ , Green  $A=0.256$ . The calculated MS is divided by the sample thickness in each case.

It follows that to a good approximation the multiple scattering can be matched to the data as described in section A10.5 to obtain the required correction. The factor  $F$  used in MSSUB should always be close to 1.

It is found that the shape of the multiple scattering is also quite insensitive to the sample geometry. Thus the procedure described in sections A10.5 should be accurate for any sample geometry and not just planar samples. An exception to this is for samples which scatter more than ~30% of the incident beam.

## Appendix 6. Format of data files produced by Analysis Programs

All files are in Ascii form;

**A. RAW, RAWB, BCORR, MSSUB, PSUBD, produce files of the form**

```

135      1189
5.000000  0.3378124  8.5653573E-02
5.500000  0.4921608  0.1176118
6.000000  0.1984558  0.1180097
.....
597.5000  -1.2792587E-02  1.3943858E-02
598.0000  1.0959029E-02  1.3809922E-02
598.5000  -1.0032296E-02  1.3844969E-02
599.0000  -3.1795382E-02  1.3847820E-02
136      1189
5.000000  0.5859909  9.3891762E-02
5.500000  1.161842  0.1287399
6.000000  0.7143860  0.1296356
.....

```

135 is the spectrum number, 1189 is the number of points

Then time, y value, error

The file list all detectors specified in the input of RAW, RAWB etc sequentially.

**B. TFIT on eg zh.dat produces a file zhd.dat which is read by TFITPLOT. This file has the format**

```

135      1024      3
50.00000  6.9565773E-03  2.6512034E-02  5.8729056E-05  9.8426826E-06
2.8947456E-05  1.9938918E-05
.....

```



135 is the detector number, 1024 the number of points in the time of flight spectra, 3 is the number of peaks fitted.

50.000 is the time of flight

6.9565773E-03 the data value

2.6512034E-02 the error on the data value

5.8729056E-05 is the fit to the data

9.8426826E-06 is the contribution of peak 1

2.8947456E-05 the contribution of peak 2

1.9938918E-05 the contribution of peak 3.

If there are more than 3 peaks fitted more values will appear in the output file.

The format is repeated for all detectors specified and for all times of flight within the ranges specified by the input to TFIT.

**C. TFIT on eg zh.dat also produces a file zhp.dat which is read by PARMEAN and PARPLOT.**

This file has the format

```

3
48

1.007900

      WEIGHT      ERROR      SIGMA      ERROR      CHI-SQ
135  9.2709E-01  2.1564E-03  4.0329E+00  6.3335E-02  1.0488E+00
136  9.2489E-01  2.0456E-03  4.1498E+00  5.8670E-02  1.0731E+00
137  9.2752E-01  2.0955E-03  3.9757E+00  5.4829E-02  1.0260E+00
.....
181  9.3174E-01  1.7427E-03  4.1767E+00  4.4978E-02  1.1171E+00
182  9.3079E-01  1.8250E-03  4.0445E+00  3.9188E-02  1.1004E+00

27.00000

      WEIGHT      ERROR      SIGMA      ERROR      CHI-SQ
135  2.6190E-02  7.7459E-04  1.1025E+01  1.0000E-06  1.0488E+00
136  2.6981E-02  7.3480E-04  1.1025E+01  1.0000E-06  1.0731E+00
.....

```

3 is the number of masses, 48 the number of detectors included. 1.0079 is the mass associated with peak 1, 27 with peak 2 etc. The program lists the values of weight and width with errors for each peak and the reduced chi-square of the fit for each detector.

#### D. Files produced by multiple scattering calculation.

These have the form;

```

3
51.00000  3.5338137E-21  6.0394530E-05  3.5847436E-06
51.00000  0.0000000E+00  1.5688147E-05  3.5265361E-06
52.00000  3.2133188E-21  2.4351232E-04  8.1685903E-06
52.00000  0.0000000E+00  1.7257749E-04  8.1390463E-06
53.00000  8.4807274E-21  3.6033263E-04  8.4261565E-08
.....

```

3 is the maximum order of scattering calculated by the program (that is neutrons which are scattered 3 times)

The first line contains

Time of flight, intensity of once scattered, intensity of twice scattered, intensity of thrice scattered

The second line contains the corresponding statistical errors given by the Monte Carlo calculation

This is repeated in steps of 1  $\mu$ sec to 562  $\mu$ sec.

## Appendix 7. Summing non-consecutive runs

If non-consecutive runs are to be summed the RAW and RAWB routines are replaced by DIFFM and DIFFMB. To run these;

First create a file containing the run numbers. For example to add runs 15103, 15120, 15115, 15118. Create a file with name e.g. run\_numbers.dat of the form

```

14188
14190
14189
14195

```

This can be done using the command "ed run\_numbers.dat" (to exit editor CONTRL Z) or using the NEDIT command.

Then for back scattering;

```
eVS> diffmb
```

```
name of file containing run numbers?
```

```
run_numbers
```

```

1 14188.00
2 14190.00

```

3 14189.00

4 14195.00

first and last spectrum numbers?

3 134

for single difference type 1

for double difference type 2

2

enter file name for output

tempb

For forward scattering;

eVS> diffm

name of file containing run numbers?

run\_numbers

1 14188.00

2 14190.00

3 14189.00

4 14195.00

First and last spectrum numbers?

135 182

File name for output?

temp

## Appendix 8. Keeping a record of your analysis

It is suggested that a record of your analysis should be kept in a Word document as you proceed. This should contain details of the names of files you have created and want to keep. Results and plots which are produced by the analysis programs can be pasted into this document by the following procedures.

1. Open a Word document
2. To record plots which are produced by the programs;
  - (a) Click on the window containing the plot.
  - (b) Press the PrtSc key on the keyboard
  - (c) Open paint via Start, All programs and accessories. There is also a paint icon at the lower right hand side of the screen in the VESUVIO cabin which can be double clicked to open Paint.
  - (d) Click on the paint window which occurs and Ctrl V to copy the screen into paint. If necessary maximize the window size to fill the screen.
  - (e) Select the plot you wish to copy using the top right hand icon in the menu on the left.

- (f) Click on the word document, go to the appropriate point and Ctrl V to paste the selected area into the Word document.
3. To record the output of programs (eg mean values calculated) in the Word document
  - (a) Select the text you wish to copy in the VMS window which runs the programs, using the mouse.
  - (b) Click on the word document go to the relevant point in the document and Ctrl V to paste the text in.

## Appendix 9. Command file produced by CRECOM

The file ZH.COM produced by the sequence of commands in section 3 is listed below.

```
$ set def USER$DISK:[JM01]
$ RAWB ! Get Raw back scattering data. Data in TEMPB.DAT
    14188    14195

$ CREBIN ! Create .IN file for fitting back scattering
zh4188
2
27.00000
91.00000
0

$ TFIT ! Fit raw back-scattering data
tempb
200 456
zh4188    b.in
3 134

$ CREFIN ! Create .IN file for fitting forward scattering.
tempbp
zh4188
1

$ RAW ! Get raw forward scattering data. Data in TEMP.DAT
    14188    14195
135 182

$ TFIT ! Fit raw forward-scattering data
```

```
temp
50 562
zh4188      .in
135 182
```

\$ CRECIN ! Create .IN file for gamma background correction

```
tempp
zh4188
```

\$ BCORR ! Correct forward scattering data for gamma background.

```
temp
zh4188      c.in
0
```

\$ TFIT ! Fit gamma corrected forward-scattering data

```
tempc
50 562
zh4188      .in
135 182
```

\$ CREMSIN ! Creates MS .IN file

```
tempcp
zh4188
      1
```

\$ THICK ! Calculates thickness parameter

```
zh4188
0.8310000
```

\$ MSCALC ! calculates multiple scattering

```
zh4188
```

\$ MSSUBB ! Subtract multiple scattering at back scattering

```
tempb
zh4188
```

\$ MSSUBB ! Subtract multiple scattering at forward scattering

```
tempc
zh4188
```

\$ PSUBB ! Obtain final forward scattering data.

```
50 562
tempcms
zh4188      .in
zh4188
135 182
278.0000    292.0000
0 0
```

\$ PSUBB ! Obtain final back scattering data.

```
50 562
tempbms
zh4188      b.in
zh4188      b
3 134
0 0
```

\$ TFIT ! Fit final back-scattering data

```
zh4188      b
200 456
zh4188      b.in
3 134
```

\$ CREFIN ! Create new .IN file for fitting forward scattering.

```
zh4188      b p
zh4188
1
```

\$ TFIT ! Fit final forward-scattering data

```
zh4188
50 562
zh4188      .in
135 182
```

\$ @EVS\$DISK0:[EVSMGR.USERPROGS]DELFILES ! Delete temporary files.

## Appendix 10. Data correction procedures

In this appendix the procedures used to correct the data for multiple scattering and gamma background are given in more detail. By following the sequence given here the user can reproduce the procedure in the command file created by the CRECOM command (see section 3) by hand and see how the different corrections affect the data.

### A10.1 Fitting the raw data

The first stage of the data analysis is to fit the raw data using the command TFIT. The fitting function which is used in this routine is described in Appendix 2. The file ZHB.IN for fitting the back scattering data is produced using the CREBIN command, as described in section 4a. The raw data TEMPB.DAT at back scattering produced by the RAWB command (see section 2) is then fitted using the TFIT command with the input file ZHB.IN.

```
eVS> tfit
```

```
Name of file containing time of flight data?
```

```
Tempb
```

This is the name of the data file produced by the RAWB routine

```
input file is tempb
```

```
tmin,tmax? (tmax-tmin must be 2**n)
```

```
300 428
```

This range was chosen by observation of the data shown in Figure 3. It includes the peaks from the masses other than mass 1 (or 2) in the data, but excludes as much as possible of the background (mainly to multiple scattering) at shorter times. For most samples this is a good range to choose. For samples containing masses <7 the range may have to be extended to include all peaks: for example 200-456  $\mu$ sec.

```
number of points= 256
```

The number of points is  $2^8$  in this case.

```
ndmin= 3 ndmax= 134
```

```
Name of file containing fit parameters?
```

```
zhb.in
```

This is the file produced by the CREBIN command.

```
npeaks= 2
```

```
1 am= 27.00000 xs= 0.0000000E+00 wid= 0.0000000E+00
```

```
2 am= 91.00000 xs= 0.0000000E+00 wid= 0.0000000E+00
```

```
FSE NOT SUBTRACTED
```

```
First and last spectrum number?
```

```
3 134
```

These are the first and last spectra to be fitted

The routine fits the data in spectra 3-134 and produces two output files. These files have the same name as the input data file with the letter D or P added. In the example these are;

(3) TEMPBD.DAT- This contains the data and fits

(4) TEMPBP.DAT – this contains the fitted parameters.

These are ASCII files with formats which are given in Appendix 6.

The data and fits can be displayed using the TFITPLOT command as described in section 4b. The fitted parameters can be examined using the PARMEAN and PARPLOT commands, as described in section 4c .

The raw forward scattering data is obtained using the command RAW as described in section 2a . This produces a data file TEMP.DAT. This is fitted using TFIT with the INPUT file ZH.IN created using the CREFIN command, as described in section 4d.

#### **A10.2. Correction of data for gamma background**

Currently the forward scattering detectors on VESUVIO suffer from a sample dependent gamma background. This background can be calculated from the measured data as described in reference [2]. It is first necessary to create a file characterising the sample. This is done using the command CRECIN (create \*.in file). This uses the fitted values for amplitudes and widths from the initial analysis at forward and backward angles to characterise the sample.

eVS> crecin

Name of file containing output of TFIT routines?

tempp

Number of masses= 3

first spectrum= 135 last spectrum= 182

1 ATOMIC MASS= 1.007900

Number of points included= 48

.....

Enter generic file name

zh

The program produces the file ZHC.IN (this will be xxxxc.in if you enter xxxx for generic file name)

3

1.007900 0.9310600 4.388812

27.00000 2.8320679E-02 12.78900

91.00000 4.0618815E-02 26.62300



The values of amplitudes and widths are those given by PARMEAN operating on TEMPP.DAT.

The necessary correction is applied by use of the command BCORR, with the input file ZHC.IN

```
eVS> bcorr
```

```
Name of file containing time of flight data?
```

```
temp
```

```
input file is temp
```

```
number of points= 1024
```

```
ndmin= 135 ndmax= 182
```

```
Enter name of .in file
```

```
zhc.in
```

```
E1= 4897.400
```

```
DE1L= 24.00000 DE1G= 74.00000
```

```
1 M= 1.007900 AREA= 0.9300000 SIGMA= 4.290000
```

```
2 M= 27.00000 AREA= 2.8999999E-02 SIGMA= 13.60000
```

```
3 M= 91.00000 AREA= 3.9999999E-02 SIGMA= 26.60000
```

```
135 RHOD= 0.4401759 PHID= -59.60000 ZD= 0.3359000
```

```
136 RHOD= 0.4706679 PHID= -59.70000 ZD= 0.2604000
```

```
.....  
181 RHOD= 0.4988186 PHID= 47.60000 ZD= -0.1835000
```

```
182 RHOD= 0.5257007 PHID= 47.50000 ZD= -0.1081000
```

The program takes about 5 minutes to run. When it has finished, the data corrected for gamma background is in the file TEMPC.DAT. The program allows the uncorrected data, the corrected data and the gamma background correction to be displayed.

```
Plot input data, corrected data and background
```

```
Plot individual detectors (1) or detector banks (2)?
```

```
type 0 to exit program
```

```
2
```

```
bank number (0 to end)
```

```
1. 135-142 2. 143-150 3. 151-158 4. 159-166
```

```
5. 167-174 6. 175-182 7. 183-190 8. 191-198
```

There are 8 banks of detectors at forward angles (see Figure A1.1).

```
1
```

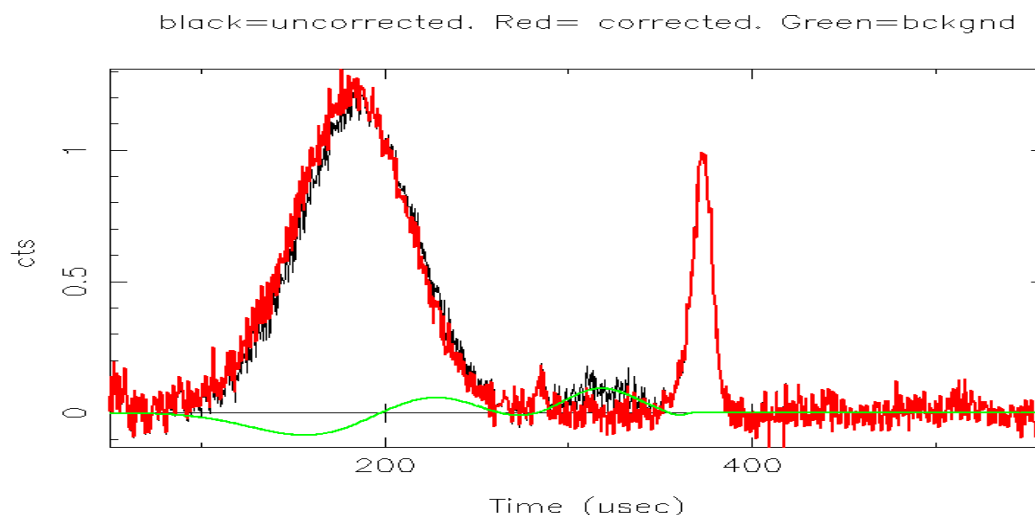


Figure A10.6. shows the uncorrected data (black), corrected data (red) and the gamma back ground (green) subtracted from the data for bank 1 (detectors 135-142).

### A10.3. Calculation of multiple scattering

The data must also be corrected for multiple scattering (MS) to get accurate values for the widths and amplitudes of the peaks. MS is calculated using Monte-Carlo methods [6]. The parameters input to the calculation are determined from the data. The first step is to fit the forward scattering data after correction for gamma background.

#### A10.3a Fit to gamma corrected data

eVS> tfit

Name of file containing time of flight data?

tempc

input file is tempc

tmin,tmax? (tmax-tmin must be 2\*\*n)

50 562

number of points= 1024

ndmin= 135 ndmax= 198

Name of file containing fit parameters?

zh.in

This is the file created using the CREFIN command (section 4d) containing the fixed values obtained from the back scattering data.

npeaks= 3

1 am= 1.007900 xs= 0.0000000E+00 wid= 0.0000000E+00

2 am= 16.00000 xs= 0.4370540 wid= 8.548523

3 am= 27.00000 xs= 0.5629457 wid= 12.64197

FSE SUBTRACTED

First and last spectrum number?

135 182

### A10.3b Creation of file for calculation of multiple scattering.

To create a file for calculating the multiple scattering use the command CREMS (create MS file). The program calculates the mean values of the parameters obtained from fitting TEMPC.DAT using PARMEAN and inserts these into the multiple scattering file.

First determine which directory you are working in using the command "sh def". For example

```
eVS> sh def
USER$DISK:[JM01]
```

Next,

```
eVS> cremsin
```

Name of file containing output of TFIT routines?

```
Tempcp
```

```
Number of masses=      3
first spectrum=    135 last spectrum=    182
      1 ATOMIC MASS=  1.007900
Number of points included=    48
wtd mean area=  0.9310600    +-  3.2063187E-04
mean area   =  0.9308982    st dev=  4.4463281E-04
```

```
Number of points included=    32
wtd mean width=  4.388812    +-  9.8748179E-03
mean width   =  4.364275    st dev=  1.7645935E-02
```

Enter generic file name

```
Zh
```

quick option (1) or slow option (2)

quick option runs 10 times faster but has larger errors. Quick option is OK for most data

```
1
```

This sequence of command creates the file ZHMS.IN given below. If in response to the prompt "Enter generic file name" you typed XXXX the file will be "XXXXms.com".

```
91 182
      3
1.007900  0.9301589  4.245532
27.00000  2.9496644E-02  13.14000
91.00000  4.0345035E-02  26.64100
241
```

1

Zh

The different lines have the following functions.

**91 182** are the first and last detectors for which the calculation is made. The back scattering detectors are arranged in three identical wedges (see Appendix 1). Hence the multiple scattering calculated in spectra 91-134 can also be used for spectra 3-90.

**3** is the number of different masses in the sample+container.

The next three lines contain the values of mass, amplitude and width of the data peak for each mass in the sample. These values are the mean values obtained via PARMEAN from the file TEMPCP.dat produced in section A10.4a by fitting the forward angle data after correction for the gamma background.

**241** is a "thickness parameter". This determines the amount of multiple scattering and is determined from the measured sample+can transmission as follows.

eVS> **thick**

Generic file name?

**zh**

sample+can transmission?

**0.831**

Measured sample+can transmission= 0.8310000

This is the sample+can transmission determined using the TRANS command described in section 3.

```
First spectrum=      91 last spectrum=      182
M=  1.007900  XSECT= 0.9301589  SIG=  4.245532
M=  27.00000  XSECT= 2.9496644E-02 SIG=  13.14000
M=  91.00000  XSECT= 4.0345035E-02 SIG=  26.64100
```

```
input thickness= 241.0000
required thickness parameter= 244.1994
required thickness parameter= 244.4864
required thickness parameter= 244.5121
required thickness parameter= 244.5145
required thickness parameter= 244.5146
required thickness parameter= 244.5146
required thickness parameter= 244.5146
```

```
required thickness parameter= 244.5146
required thickness parameter= 244.5146
required thickness parameter= 244.5146
NAME OF OUTPUT FILE IS zh      MS.IN
```

The routine THICK produces a new file ZHMS.IN with the thickness parameter adjusted to give the same transmission as that measured (0.831) in this case. The multiple scattering correction can be run interactively by the command MSCALC.

```
eVS> mscalc
```

```
Instrument parameters read from fileEVS$DISK0:[EVSMGR.CALIB.PAR]IP0004
```

```
Generic filename?
```

```
zh
```

```
Input file iszh      ms.in
First spectrum=      91 last spectrum=      182
TMIN= 50.00000    TMAX= 562.0000    DT= 1.000000
M= 1.007900    XSECT= 0.9301589    SIG= 4.245532
M= 27.00000    XSECT= 2.9496644E-02 SIG= 13.14000
M= 91.00000    XSECT= 4.0345035E-02 SIG= 26.64100
```

```
TRANSMISSION= 0.8310000    ATTENUATION= 0.1690000
First spectrum=      3 last spectrum=      182
TMIN= 50.00000    TMAX= 562.0000    DT= 1.000000
  1 M= 1.007900    XSECT= 0.9300000    SIG= 4.260000
  2 M= 27.00000    XSECT= 2.8999999E-02 SIG= 13.50000
  3 M= 91.00000    XSECT= 3.9999999E-02 SIG= 26.60000
TRANSMISSION= 0.8598490    ATTENUATION = 0.140151
ENTER NAME OF OUTPUT FILE
NAME OF OUTPUT FILE IS zhms
```

The program will start to run and will print out the above lines. As can be seen from the output of the program, the calculated sample transmission with the sample thickness calculated by the THICK command was equal to the measured transmission of 0.83. The program typically takes ~10 minutes run on the fast option and ~100 minutes on the slow option. After the program has completed, the calculated multiple scattering is contained in the files ZH91.dat ...ZH182.DAT.

## A10.4. Correction of data for multiple scattering.

### A10.4a Backscattering

When the MS calculation has completed, the back scattering data can be corrected for multiple scattering using the command MSSUB.

```
eVS> mssub
```

```
Name of file containing time of flight data?
```

```
tempb
```

```
Name of input file =tempb
```

This is the uncorrected back-scattering data produced by RAWB.

```
first spectrum=      3 last spectrum=    134
```

```
dt= 0.5000000
```

```
tmin= 5.000000    tmax= 599.0000
```

```
Name of file containing ms calculation?
```

```
zh
```

This is the generic file name

```
Name of file containing ms calculation =[evsmgr.jm.ms]zhms
```

The program plots the data as a black line, the calculated total scattering as a red line and the calculated multiple scattering as a green line. The scale of both data and calculated multiple scattering is arbitrary and the multiple scattering is normalised so that the number of counts in data and calculation is the same within the region 320- 400  $\mu$ sec.

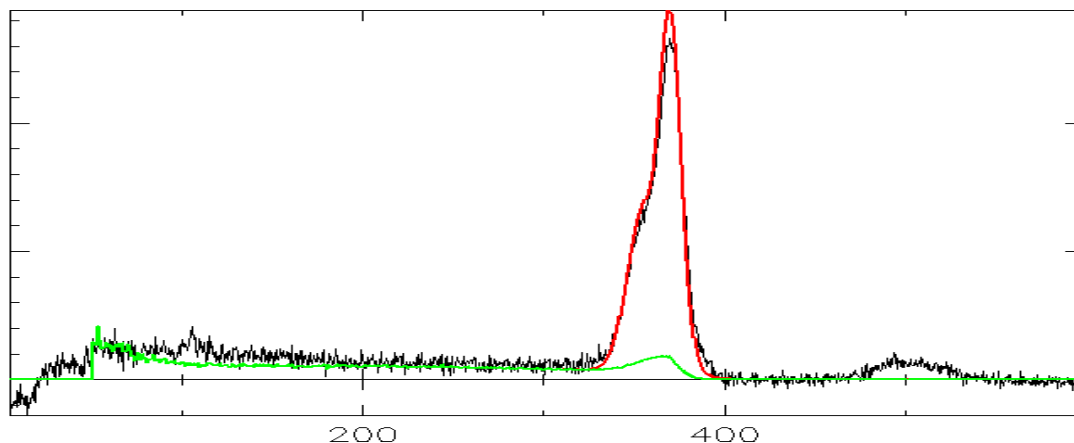


Figure A10.7. Plot produced by MSSUB. The black line is the data. The green line is the calculated multiple scattering. The red line is the calculated total scattering. This coincides with the green line to the left of the aluminium peak and cannot be seen

As discussed in Appendix 5 the shape of the multiple scattering contribution is insensitive to the sample geometry and thickness, providing the sample is not too thick (30% scattering is the maximum which can be measured, although 10% scattering is recommended). Only the ratio of single/multiple scattering varies significantly with thickness and sample geometry. The procedure described here can therefore be used to correct any sample geometry for multiple scattering to a good approximation.

MSSUB now gives the prompt

factor by which MS is multiplied? (0 TO EXIT)

suggested factor=1.02145

1

The program allows the multiple scattering to be multiplied by a constant factor  $F$  and plots the data after subtraction of the multiple scattering multiplied by  $F$ . The program calculates a suggested factor by integrating calculation and data for the back-scattering data over the range 200-280  $\mu\text{sec}$ . The factor is chosen automatically when the command file produced by CRECOM (section 3) is run.

In this case it can be seen that with  $F=1$  the calculated multiple scattering correction closely matches that observed in the data region of interest. This is inferred from the fact that the background to the left of the aluminium and zirconium peaks, which can be observed in Figure A10.7, has been removed in fig A10.8. For flat planar samples covering the neutron beam,  $F$  should be close to 1. However with more complex sample geometries  $F \neq 1$  may be necessary. In the latter case various factors can be tried to give the best zero base line to the left of the peaks from the heavier atoms.

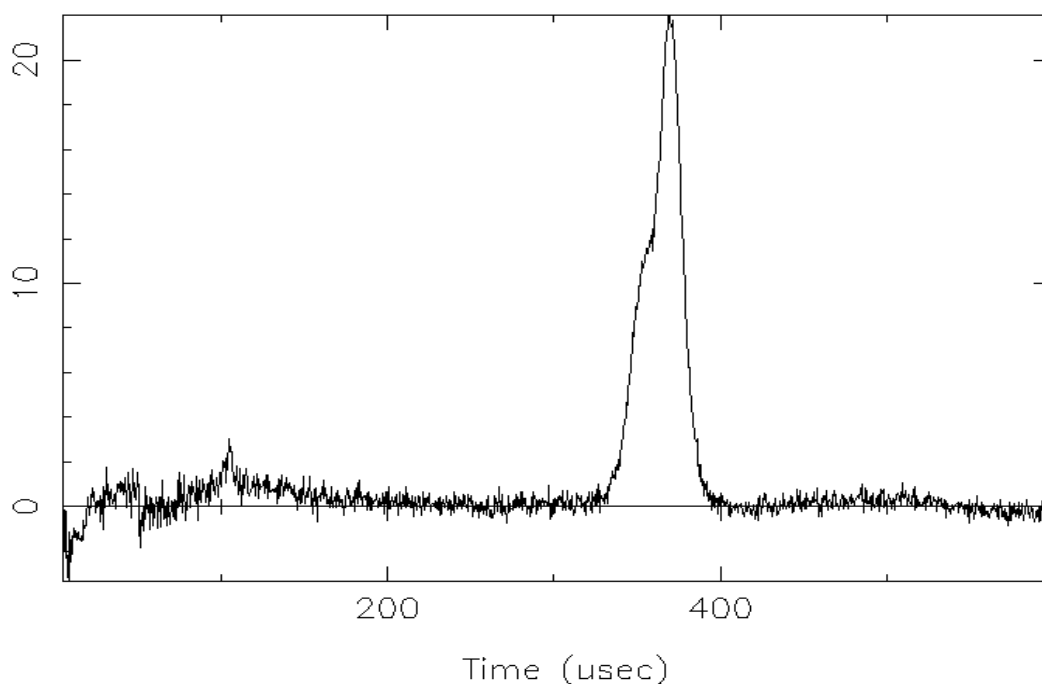


Figure A10.8. Plot produced by MSSUB of data after subtraction of multiple scattering with  $F=1$ .

Once  $F$  has been determined, type 0 in response to the prompt.

factor by which MS is multiplied? (0 TO EXIT)

0

The program then asks for the final factor by which the calculated multiple scattering is to be multiplied and the file name for output.

factor by which MS is multiplied for output file?

1

Name of output file =tempms.dat

The corrected back scattering data is now contained within the file TEMPMS.DAT. It can be viewed using the routine PLOTD. If you wish to keep this file for a more accurate determination of the amplitudes and widths at back-scattering it should be renamed. For example

eVS> Rename TEMPMS.DAT ZHB.DAT

However this file can be quickly regenerated using the RAWB and MSSUB routines and it is usually not necessary to keep it.

#### A10.4b Forward scattering

The multiple scattering correction for forward scattering is made by an essentially identical procedure to that used for the back-scattering data.

eVS> mssub

Name of file containing time of flight data?

tempc

This is the forward angle data after correction for gamma background

Name of input file =tempc

first spectrum= 135 last spectrum= 182

dt= 0.5000000

tmin= 50.00000 tmax= 561.5000

Name of file containing ms calculation?

zh

Name of file containing ms calculation =[evsmgr.jm.ms]zh

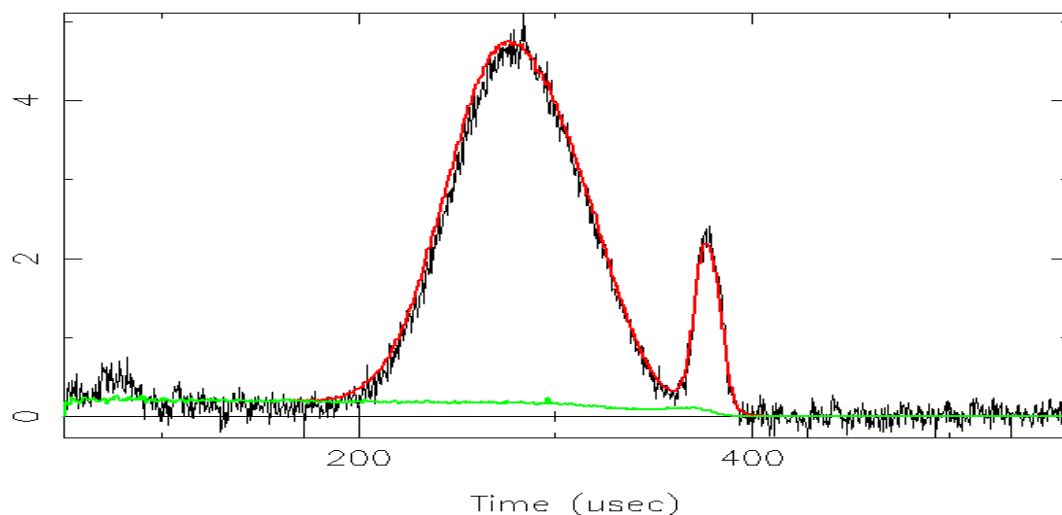




Figure A10.9. Plot produced by MSSUB. The black line is the sum of all data in the input data file with angles between 30 and 50 degrees.. The red line is the calculated total scattering. The green line is the multiple scattering contribution.

The program plots the sum of the data in the angular range 30-50 degrees as the black line, the sum of the calculated scattering as the red line and the calculated multiple scattering as the green line. The range 30-50 degrees is chosen for this plot as it contains a flat background between 120 and 180  $\mu\text{sec}$ , which can be used to assess the accuracy of the calculation of multiple scattering. For forward scattering the suggested factor is calculated by integrating data and calculation for angles between 30 and 40 degrees, over the range 120-180  $\mu\text{sec}$ .

factor by which MS is multiplied? (0 TO EXIT)

suggested factor=0.97218

0.972

The program displays the data after subtraction of the multiple scattering with the chosen factor. This factor should be close to one. However it is chosen to give the best zero background in the range 120-180  $\mu\text{sec}$ .

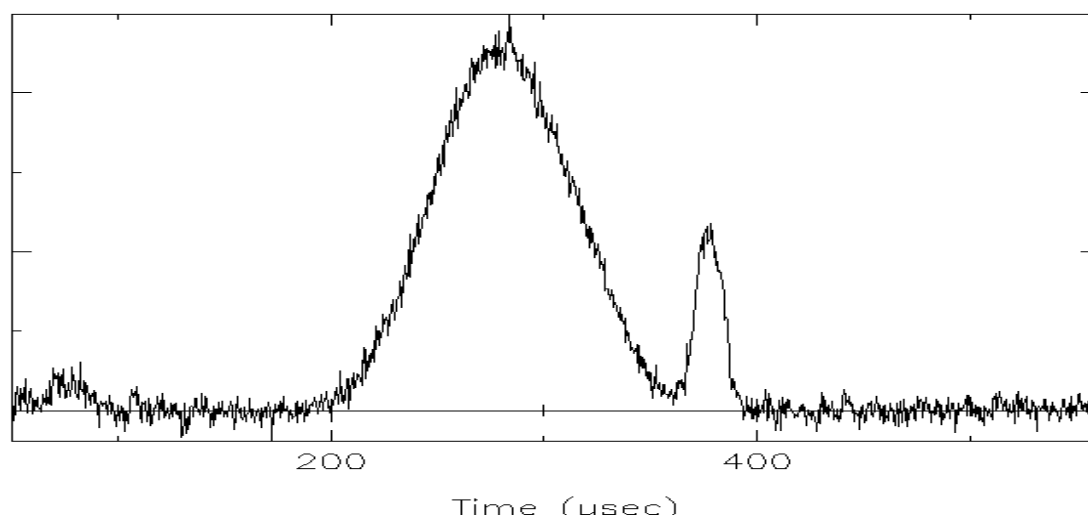


Fig A10.10. Plot produced by MSSUB of data – MS with the chosen factor  $F=1$ . The structure which can be observed for  $t < 120 \mu\text{sec}$  is due to a second gold resonance in the analyser foil at energy  $E \sim 60 \text{ eV}$ . This region is removed from the final data set using PSUBD (see section A10.5c).

factor by which MS is multiplied? (0 TO EXIT)

0

If 0 is not entered the program re-plots with the new factor. If 0 then the program goes to the final step for creating the data file with MS subtracted

factor by which MS is multiplied for output file?

0.972

Name of output file =tempms

The program writes the data corrected for multiple scattering to the file TEMPMS.DAT.

#### A10.4c Removal of spurious contributions to the data.

The final step in the data correction procedure is to subtract;

- (1) The contribution from a 60 eV resonance in the gold analyser. This occurs at short times (<120  $\mu\text{sec}$ ) and must also be removed from the data set. Its effects can be seen in Figure A10.10, where it produces structure at times below 120  $\mu\text{sec}$ . This structure mimics that produced by the 4.9 eV resonance, but with lower intensity.
- (2) Some samples with heavy elements give small nuclear resonance peaks over narrow regions of the time of flight spectra. One example is the peak at ~280  $\mu\text{sec}$  due to hafnium impurities in the  $\text{ZrH}_2$  sample which can be observed in figure 4. Such regions must also be removed from the data.

Both subtractions are performed by the command PSUBD.

```
eVS> psubd
```

```
tmin,tmax? (tmax-tmin must be 2**n)
```

```
50 562
```

```
Name of file containing time of flight data?
```

```
tempms
```

```
Name of file containing fit parameters?
```

```
zh.in
```

```
input file is tempms
```

```
Name of output file?
```

```
zh
```

```
This is your generic file name
```

```
number of points= 1024
```

```
ndmin= 135 ndmax= 182
```

```
First and last spectrum number?
```

```
135 182
```

```
enter tmin,tmax for data to be excluded
```

```
to exit enter tmin=tmax
```

```
278 292
```

The region 278-292  $\mu\text{sec}$  is excluded as it contains a contribution from hafnium impurities in the sample. This can be seen for example in Figure 4.

```
enter tmin,tmax for data to be excluded
```

```
to exit enter tmin=tmax
```

```
0 0
```

Only one data region is excluded by hand in this case. The program next prompts the user to display the data after elimination of spurious data.

```
plot individual detectors (1) or banks (2)?
```

2

bank number (0 to end)

1. 135-142 2. 143-150 3. 151-158 4. 159-166  
5. 167-174 6. 175-182 7. 183-190 8. 191-182

1

This produces the plot shown in Figure A10.11. Two regions have been eliminated from the data set; (i) A region between 100 and 116  $\mu\text{sec}$  is contaminated by the second gold resonance and is automatically removed. (ii) The region 278-292  $\mu\text{sec}$  was excluded by hand in response to the user input to the program. These regions appear as gaps in the plotted spectrum. The regions which are eliminated from the data are given very large error bars so that they do not affect any fits. The final data set is stored in the file ZH.DAT (XXXX.DAT if your generic file name is XXXX).

1

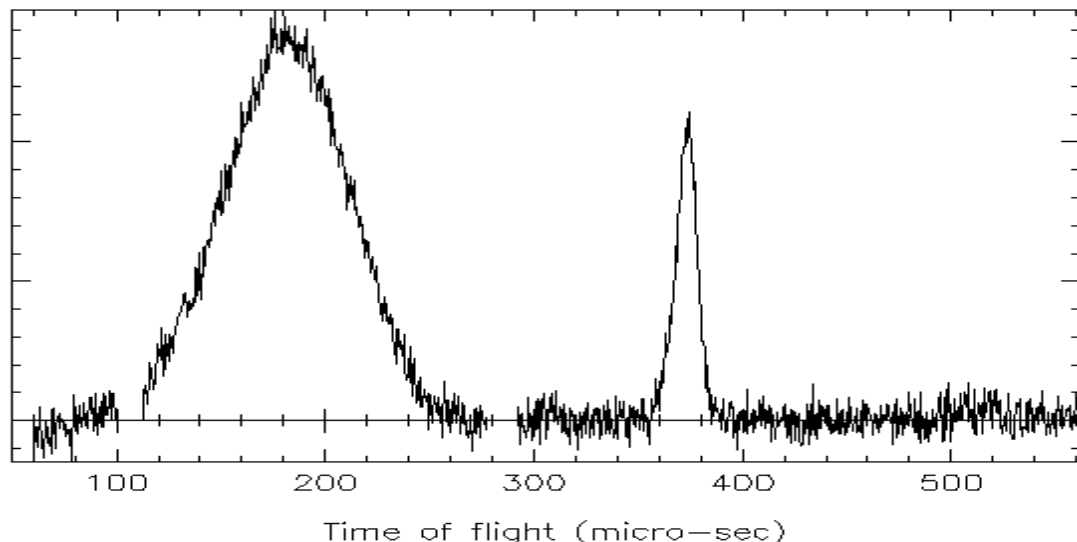


Figure A10.11. Shows the final data set from bank 1 (sum of spectra 135-142) as the black line. The gaps are data regions eliminated by PSUBD.

This completes the data correction procedures. In principle one should now repeat the procedure, using the widths and amplitudes obtained from the corrected data to produce the file ZHC.IN for calculation of the gamma correction and ZHMS.COM for the calculation of multiple scattering. However it is found empirically that this is not necessary, since the gamma correction and multiple scattering correction are both insensitive to the exact values of amplitudes and widths of the peaks. Hence iteration makes no significant difference to the final results obtained.

It is recommended that once the final corrected data sets at forward (ZH.dat) and backward (ZHB.dat) have been obtained all data files produced by the intermediate steps are deleted. Otherwise you will run out of disk space quite rapidly. This can be done by the command

eVS> delete temp\*.dat;\*

If necessary all TEMP files can be recreated quickly by rerunning the programs with the relevant .IN files. The latter should not be deleted.

---

## References

- 1 *Calibration of the electron volt spectrometer, a deep inelastic neutron scattering spectrometer at the ISIS pulsed neutron spallation source.* A. L. Fielding and J. Mayers, *Nucl. Inst. Meth. A*, **480**, 680 (2002).
- 2 *Calculation of background effects on the VESUVIO eV spectrometer*, J Mayers Rutherford-Appleton :Laboratory Technical Report RAL-TR-2010-012. Submitted to *Nuc Inst and Meth*.
- 3 *Calibration of the electron volt spectrometer VESUVIO at ISIS.* Rutherford-Appleton Laboratory technical report, J Mayers and M A Adams, RAL-TR-2009-22. Submitted to *Measurement Science and Technology*
- 4 *Double Difference method to improve the resolution of an eV neutron spectrometer.* P.A. Seeger, A.D. Taylor and R. M. Brugger, *Nucl. Instr. Methods A* **240**, 98 (1985).
- 5 *Foil cycling technique for the VESUVIO spectrometer operating in the resonance detector configuration*, E M Schoonveld, J. Mayers et al *Rev. Sci. Inst.* **77** 95103 (2006)  
Author(s): Schooneveld EM, Mayers J, Rhodes NJ, et al.
- 6 J. Mayers, A.L. Fielding and R. Senesi, *Nucl. Inst. Methods A* **481**, 454 (2002)
- 7 *Data Analysis. A Bayesian Tutorial*, D. Sivia (Clarendon Press Oxford 1996) , Section 4.2.
- 8 *Measurement of momentum distributions of light atoms and molecules in condensed matter systems using inelastic neutron scattering.* C Andreani, D Colognesi, J Mayers and G Reiter. *Adv Phys* **54** 377-469 (2005)
- 9 *Excess of proton mean kinetic energy in supercooled water.* A Pietropaolo, R Senesi, C Andreani et al. *Phys. Rev. Lett.* **100** 127802 (2008).
- 10 *Proton momentum distribution in liquid water from room temperature to the supercritical phase.* C. Pantalei, A. Pietropaolo, R. Senesi, S. Imberti, C. Andreani, J. Mayers, C. Burnham, and G. Reiter. *Phys. Rev. Lett.* **100** 177801 (2008)
- 11 *Measurement of the kinetic energy and lattice constant in hcp solid helium at temperatures 0.07-0.4K.* M Adams and J Mayers *Phys. Rev. Lett.* **98** 85301(2007).
- 12 *Quantum and classical relaxation in the proton glass .* Y. Feng, C. Ancona-Torres et al. *Phys. Rev. Lett.* **97** 69901 (2006)
- 13 *Anomalous behaviour of proton zero point motion in water confined in carbon nano-tubes.* G. Reiter, C. Burnham et al *Phys. Rev. Lett.* **97** 247801 (2006).
- 14 *Proton momentum distribution in a protein hydration shell.* R. Senesi, A Pietropaolo et al. *Phys. Rev. Lett.* **98** 138102 (2007)

- 
- 15 *Measurement of the 3-D Born-Oppenheimer potential of a proton in a hydrogen-bonded system via deep inelastic neutron scattering.* D Homouz, G. Reiter et al. *Phys. Rev. Lett* **98** 115502 (2007)
  - 16 *The measurement of anomalous neutron inelastic cross-sections at electronvolt energy transfers.* J Mayers and T Abdul-Redah, *J. Phys. Condens. Matter* **16** (2004) 4811–4832
  - 17 *Pulsed Neutron Scattering*, C. Windsor, Taylor and Francis, London, (1821).
  - 18 *Advances on detectors for low-angle scattering of epithermal neutrons* E P Cippo, G Gorini, M Tardocchi, et al. *Measurement Science and Technology* **19** 47001 (2008)
  - 19 *YAP Scintillators for resonant detection of epithermal neutrons at pulse neutron sources.* M. Tardocchi, G. Gorini et al. *Rev. Sci. Inst.* **75**, 4880 (2004).
  - 20 *A resonant detector for high-energy inelastic neutron scattering experiments.* C. Andreani, A. Pietropaolo et al. *Applied Physics Letters* **85** 5454 (2004)
  - 21 *Foil cycling technique for the VESUVIO spectrometer operating in the resonance detector configuration*, E M Schoonveld, J. Mayers et al *Rev. Sci. Inst.* **77** 95103 (2006) Schooneveld EM, Mayers J, Rhodes NJ, et al.
  22. S.W. Lovesey, *Theory of Neutron Scattering from Condensed Matter*, Vol 1, (Oxford University Press, New York 1824).
  - 23 V. F. Sears *Phys. Rev. B.* **30**, 44 (1984).
  - 24 J Mayers, C Andreani, G Baciocco, *Phys Rev B* **39** 2022 (1989)
  - 25 J Mayers *Phys Rev B* **41**, 41 (1990)
  - 26 See H R Glyde *Phys Rev B.* **50** 6726, (1994) for a recent review of the literature on FSE.
  27. A.C. Evans, D.N. Timms, J. Mayers and S. Bennington, *Phys Rev B* **53** 3023 (1996).
  - 28 A L Fielding, D.N. Timms, A.C. Evans and J. Mayers *J. Phys. Condens. Matter* **8** 7205 (1996)
  - 29 G. Reiter and R. Silver, *Phys. Rev. Lett.* **54**, 1047 (1825).
  - 30 G. F. Reiter, J. Mayers, and J. Noreland, *Phys. Rev. B* **65**,104305 (2002).