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TITLE: ANALYSIS OF INELASTIC NEUTRON SCATTERING SPECTRA FROM
A TIME-OF-FLIGHT SPECTROMETER WITH FILTER DETECTOR

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Analysis of Inelastic Neutron Scattering Spectra from a Time-of-Flight Spectrometer with Filter Detector

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Abstract. Inelastic neutron scattering spectra obtained from time-of-flight spectrometers with filter detector suffer in energy resolution from a long time-of-flight tail in the filter response function. A mathematical method is described which removes this tail in measured spectra. The energy resolution can thereby be adapted for each part of the spectrum. Applications of the method to data taken at the LANSCE pulsed spallation source are presented.

1. Introduction

Polycrystalline filters can be used as energy analysers ("filter detector") of the scattered neutrons in inelastic neutron scattering experiments (Brockhouse et al 1960). In spectrometers based on this method the detector may present a large solid angle to the scattering sample, which will result in a high intensity of detected neutrons. Detected neutrons are those scattered by the sample with energies within the filter bandpass. For the most common polycrystalline filter materials, beryllium and beryllium oxide, this bandpass is 0 - 5.22 meV and 0 - 3.76 meV, respectively.

To obtain the scattering function of the sample the incident neutron energy must be scanned. Using the "white" incident beam of a pulsed neutron source this scanning can be performed by time-of-flight (TOF) methods. However, a TOF spectrometer measures the total flight time $t=t_1+t_2$ from the moderator to the sample (t_1) and from there to the detector (t_2). As the filter bandpass is poorly defined on the time-of-flight scale for the filter materials mentioned above (theoretically $1000 \mu\text{s/m} \leq t_2 < \infty$ for Be and $1180 \mu\text{s/m} \leq t_2 < \infty$ for BeO) so is t and, therefore, the incident neutron flight time $t_1=t-t_2$.

A filter difference method (Pan et al 1966, Taylor et al 1984) has been applied to overcome this problem. This method uses data measured separately with a Be filter and with a BeO filter. These data are then combined to reduce the effective filter bandpass to 3.76 - 5.22 meV in neutron energy and to $1000 \mu\text{s/m} \leq t_2 < 1180 \mu\text{s/m}$ in neutron time-of-flight, respectively.

The principal layout of such a spectrometer at a spallation neutron source is given in Fig. 1 which refers to the Filter Difference Spectrometer (FDS) at the Los Alamos Neutron Scattering Center (LANSCE). This spectrometer uses 10 filter segments, five of which are Be and five are BeO. They are arranged in such a way that each filter has an opposite counterpart at the same scattering angle.

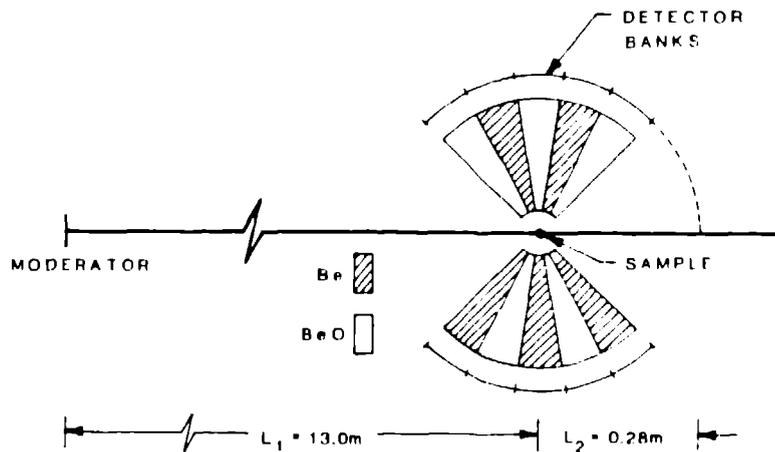


Fig. 1
Principal layout of
a filter difference
spectrometer (FDS at
LANSCE)

The filter difference method, however, still has several disadvantages:

- The use of two filter materials halves the available solid angle.
- Be and BeO filters of the same length have different (energy dependent) transmission values.
- The energy resolution of filter difference spectra is fixed by the filter difference energy bandpass and cannot be optimally selected.

These disadvantages can be removed by analysing spectra taken with one filter material only and applying a mathematical method which performs a "filter difference" based on the filter response function. Such a method is described in the following.

2. The filter response function

Neutrons scattered by a sample of cross section

$$\frac{d^2\sigma}{d\Omega dE_2} = \sqrt{\frac{E_2}{E_1}} S(Q, \hbar\omega) \quad (1)$$

are recorded as a spectral distribution in total time-of-flight $t = t_1 + t_2$ given by

$$g(t) = f(E_1) \frac{d^2\sigma}{d\Omega dE_2} T(E_2) \epsilon(E_2) \frac{dE_2}{dt} \\ = f(E_1) \sqrt{\frac{E_2}{E_1}} S(Q, \hbar\omega) T(E_2) \epsilon(E_2) \frac{dE_2}{dt} \quad (2)$$

where $\hbar\omega = E_1 - E_2$ is the energy transfer and Q the momentum transfer of the neutrons, $f(E_1)$ the distribution of incident neutron energies, $T(E_2)$ the filter transmission, and $\epsilon(E_2)$ the detector efficiency. The indices 1, 2 refer to incident and scattered neutrons, respectively. With

$$t = t_1 + t_2 = \sqrt{\frac{m}{2}} \left(\frac{L_2}{\sqrt{E_2 + \hbar\omega}} + \frac{L_1}{\sqrt{E_2}} \right) \quad (3)$$

the derivative dE_2/dt is

$$\frac{dE_2}{dt} = \frac{(2 E_1)^{3/2}}{\sqrt{m} L_1 \left[1 + \frac{L_2}{L_1} \left(\frac{E_1}{E_2} \right)^{3/2} \right]} \quad (4)$$

For a δ -function excitation in the sample at energy transfer $\hbar\omega = \hbar\omega_0$ the spectral distribution is then given by

$$g(t, \hbar\omega_0) = f(E_1) T(E_2) \varepsilon(E_2) \frac{2 E_1 \sqrt{E_2}}{\sqrt{\frac{m}{2}} L_1 \left[1 + \frac{L_2}{L_1} \left(\frac{E_1}{E_2} \right)^{3/2} \right]} \quad (5)$$

which, for convenience, can be written in neutron-time-of-flight and neutron velocity as

$$g(t, \hbar\omega_0) = f(t_1) T(v_2) \varepsilon(v_2) \frac{1}{\frac{v_1}{v_2} \left[1 + \frac{L_2}{L_1} \left(\frac{v_1}{v_2} \right)^3 \right]} \quad (6)$$

Velocity v_2 is related to velocity v_1 by $E_2 = E_1 - \hbar\omega_0$.

The spectrum (6), with the incident TOF distribution $f(t_1)$ and the detector efficiency $\varepsilon(v_2)$ taken as unity, is the filter response function $\theta(t)$. Normalized to the filter response at the upper filter bandpass energy E_c ("filter cut-off") this function is given as

$$\theta(t) = \frac{T(v_2) \frac{\tilde{v}_1}{v_c} \left[1 + \frac{L_2}{L_1} \left(\frac{\tilde{v}_1}{v_c} \right)^3 \right]}{T(v_c) \frac{v_1}{v_2} \left[1 + \frac{L_2}{L_1} \left(\frac{v_1}{v_2} \right)^3 \right]} \quad (7)$$

where \tilde{v}_1 is determined by the equation $\tilde{E}_1 = E_c + \hbar\omega_0$. This expression differs from the corresponding one given by Windsor (1981) and by Taylor et al (1984)

$$\theta^*(t_2) \propto \frac{T(v_2)}{T(v_c)} \left(\frac{v_2}{v_c} \right)^4 \quad (8)$$

which has been derived taking the differential in eq. (2) with respect to t_2 , i.e. neglecting that the measured quantity in the neutron time-of-flight experiment is the total flight time $t = t_1 + t_2$.

The filter response function (7) for various energy transfers $\hbar\omega_0$ is shown in Fig. 2 for a Be filter of length 15 cm. The filter transmission used in the computation takes into account neutron losses by absorption and inelastic scattering processes in the filter. It is given by

$$T(E_2) = \exp(-\Sigma(E_2) d) \quad \text{if } E_2 < E_c \quad (9)$$

$$= 0 \quad \text{if } E_2 > E_c$$

where d is the filter length. The macroscopic cross section $\Sigma(E_2)$ has an $1/v$ -behaviour and is temperature dependent. Numerical values at $E_2 = 1$ meV are $\Sigma = 0.0851$ cm⁻¹ for 300 K and $\Sigma = 0.0111$ cm⁻¹ for 100 K (Taylor et al 1984)

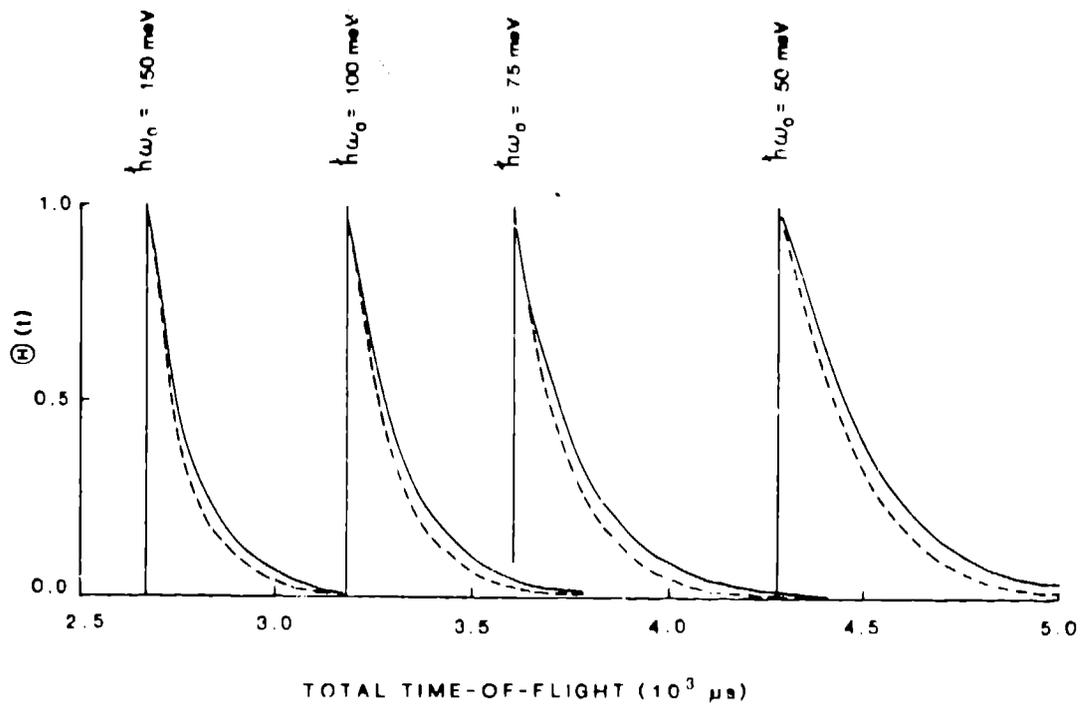


Fig. 2 Filter response function for a Be filter of length 15 cm (solid line: filter at 100 K, dashed line: filter at room temperature). The total time-of-flight refers to the flight path lengths given in Fig. 1.

As Fig. 2 shows the filter response function has a sharp leading edge and a tail which decreases with increasing total time-of-flight. The leading edge corresponds to neutrons which pass the filter with the cut-off energy E_c and whose energy before scattering thus was $\tilde{E}_1 = E_c + \hbar\omega_0$. These neutrons are, for a given $\hbar\omega_0$, those recorded with the shortest total time-of-flight. The length of the time-of-flight tail depends on the energy transfer $\hbar\omega_0$ such that smaller energy transfers lead to longer tails. This property, which is not predicted by (8), reflects that the fixed bandpass energy $0 - E_c$ gives a longer tail on the total time-of-flight scale for lower incident energies. In the elastic limit ($\hbar\omega_0 = 0$) the response function (7) reduces to

$$\theta(t) = \frac{T(v_2)}{T(v_c)} \quad (10)$$

i.e. besides an attenuation by the filter transmission the time-of-flight tail extends to infinity. (Note, however, that the measured spectra are multiplied by the incident spectrum $f(t_1)$ which decreases for large time-of-flights, i.e. long wavelengths.)

3. The method of lineshape analysis

Equation (6), which was derived for the lineshape of a δ function excitation, can be applied to measured TOF spectra to subtract the time-of-flight tail produced by the filter response function. This method starts with the first channel in the measured (background corrected) spectrum (channels $1 \geq i \geq N$) which has intensity $S(i) > 0$. This intensity corresponds to an excitation recorded at the leading

edge of the filter response function, i.e. for this TOF spectrum channel i holds the relation

$$\hbar\omega_0(i) = \tilde{E}_1(i) - E_c \quad (11)$$

where the incident energy $\tilde{E}_1(i)$ is determined by the time-of-flight relation

$$\tilde{t}_1(i) = t(j) - t_c \quad (12)$$

The time-of-flight tail in the following TOF channels $j=i+1, \dots, N$ due to the excitation at $\hbar\omega_0(i)$ is given by

$$s(i, j) = S(j) \frac{g(t(j), \hbar\omega_0(i))}{g(t(i), \hbar\omega_0(i))} \quad (13)$$

To compute $g(t(j), \hbar\omega_0(i))$ with relation (6), $t_1(j)$ and $t_2(j)$ must be known, whereas only their sum $t(j) = t_1(j) + t_2(j)$ is determined by the experimental conditions. Writing

$$t = t_1 + t_2 = t_1 + \frac{L_2}{\sqrt{\left(\frac{L_1}{t_1}\right)^2 - \frac{2}{m} \hbar\omega_c}} \quad (14)$$

t_1 as a function of t and ω_c can be obtained by solving this equation.

This is most conveniently done by an approximate iteration process using the relation

$$\frac{dt}{dt_1} = 1 + \frac{L_2}{L_1} \left(\frac{v_1}{v_2} \right)^3 \quad (15)$$

The subtraction of the time-of-flight tail (13) of the excitation at $\hbar\omega_0(i)$ transforms the measured TOF spectrum $S(j)$ in a first step to

$$S_1(j) = S(j) - s(i, j); \quad j=i+1, \dots, N \quad (16)$$

If the subtraction is successively applied for scattering intensities $\hbar\omega_0(k); k=i, \dots, N$; i.e. if the recursion

$$S_1(j) = S(j) - \sum_{k=i}^{j-1} S_1(k) \frac{g(t(j), \hbar\omega_0(k))}{g(t(k), \hbar\omega_0(k))} \quad (17)$$

is performed for all channels $j=i+1, \dots, N$, the measured spectrum $S(j)$ is transformed to a spectrum $S_1(j)$ in which the remaining intensity in each channel is corrected for the time-of-flight tail due to excitations recorded in preceding channels.

4. Resolution

Beside the filter response function (7), the resolution of a TOF spectrometer with filter detector has additional contributions from TOF uncertainties due to neutron pulse width, sample and detector dimensions and rise of the filter edge at the cut-off energy.

These TOF uncertainties can be calculated and compared with experimental results for elastic scattering ($\hbar\omega_0=0$) of 5.2 meV neutrons, using the measured edge of the Be cut-off. This comparison is made in Fig. 3 where also the single contributions to the TOF uncertainties are given. The agreement between calculation and experiment is very good.

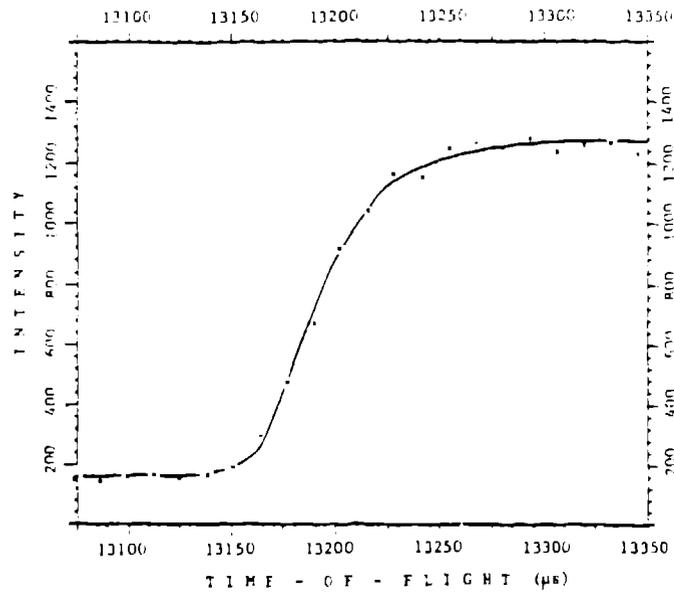


Fig. 3
Comparison of the measured Be (10 $\bar{1}$ 0) edge with the filter response function for elastic scattering, convoluted with the TOF uncertainties for 5.2 meV neutrons (refers to the FDS at LANSCE)

Resolution contributions:

- moderator pulse shape
 $M(t) = \exp(-\beta t)$; $1/\beta = 28 \mu s$ for 5.2 meV
- sample and detector dimensions
gaussian; $\sigma = 11.9 \mu s$ for 5.2 meV
- Be (10 $\bar{1}$ 0) edge, width $12.5 \mu eV \hat{=} 16 \mu s$

A calculation of these additional TOF uncertainties for energy transfers $\hbar\omega_0 > 0$ reveals that they are small compared to the effect of tail of the filter response function. Furthermore, since the filter response function for nominal flight times within these uncertainties changes insignificantly, it can thus be proved that the effective scattering function as derived by expression (17) agrees with the sample scattering function broadened by the additional TOF uncertainties (at least under the instrumental conditions of the FDS at LANSCE).

In principle, a correction for these TOF uncertainties could be performed by conventional deconvolution techniques. This, however, is not subject of this paper.

5. Applications

The method of lineshape analysis is applied to data taken with the spectrometer FDS at LANSCE. The first example is a measurement on zirconium hydride ZrH_2 with the sample at room temperature. Fig. 4 (a) gives the measured TOF spectrum with Be filters (at 100 K) in front of the detectors. Fig. 4 (b) shows the spectrum computed from (a), using relation (17) to remove the tail of the filter response function. Note that the peaks in the corrected spectrum are narrower and shifted to lower time-of-flights.

ZrH₂ (ROOM TEMP.)

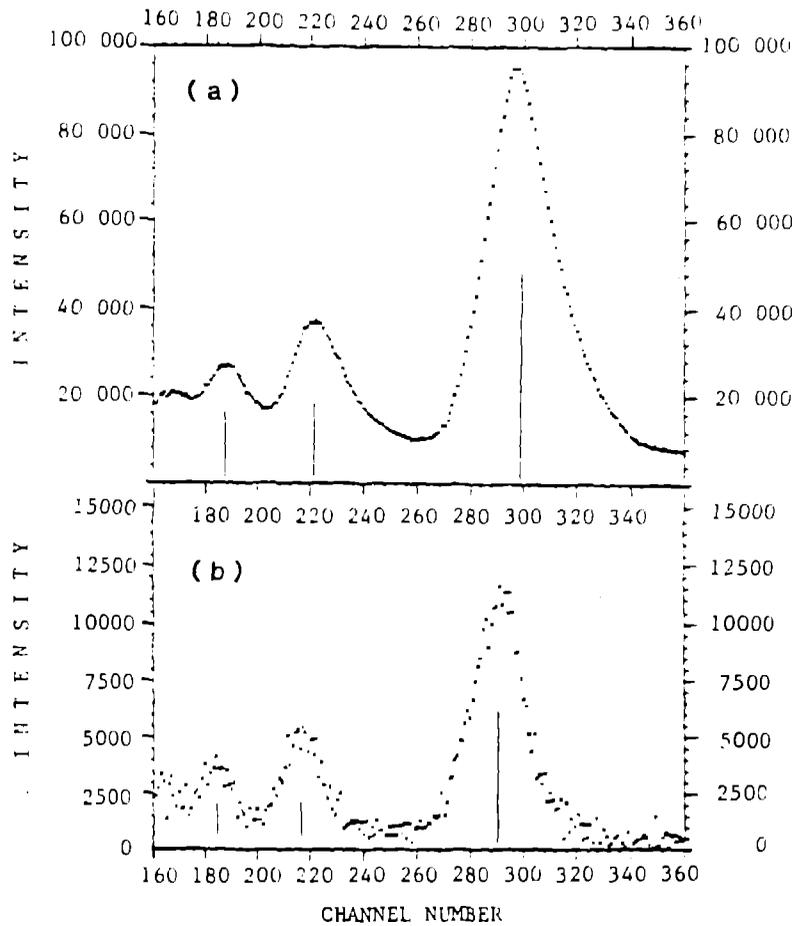


Fig. 4
Inelastic neutron scattering spectrum of optical modes in ZrH₂ at room temperature (channel width is 9.6 μ s)
(a) data taken with Be filters
(b) data corrected for the filter response function

The second example is chromous acid, CrOOH, measured at a sample temperature of 15 K. The inelastic neutron scattering spectrum of CrOOH is known to have a narrow isolated peak at energy transfer $\hbar\omega=150$ meV (Taylor et al 1984, Tomkinson et al 1985). Fig. 5 gives this part of the TOF spectrum taken (a) with the Be filters, (b) with the BeO filters and (c) after applying the conventional (Be-BeO) filter difference method. (The filters were at room temperature.)

It will be shown that this filter difference can be performed mathematically with the method described above using only spectrum (a). For this procedure a window has to be set which corresponds to the filter difference bandpass, i.e. only for flight times beyond this window the remaining tail of the filter response function has to be considered in correcting the data. Given this window is n channels, the expression for the 'filter difference spectrum' is

$$S_n(j) = S(j) - \sum_{k=i}^{j-n} S_1(k) \frac{g(t(j), \hbar\omega_0(k))}{g(t(k), \hbar\omega_0(k))} \quad (18)$$

which, for $n=1$, is identical with (17).

The transformation (18) of the data in Fig. 5 (a) results in Fig. 5 (d) which agrees quite well with the direct filter difference result of Fig. 5 (c).

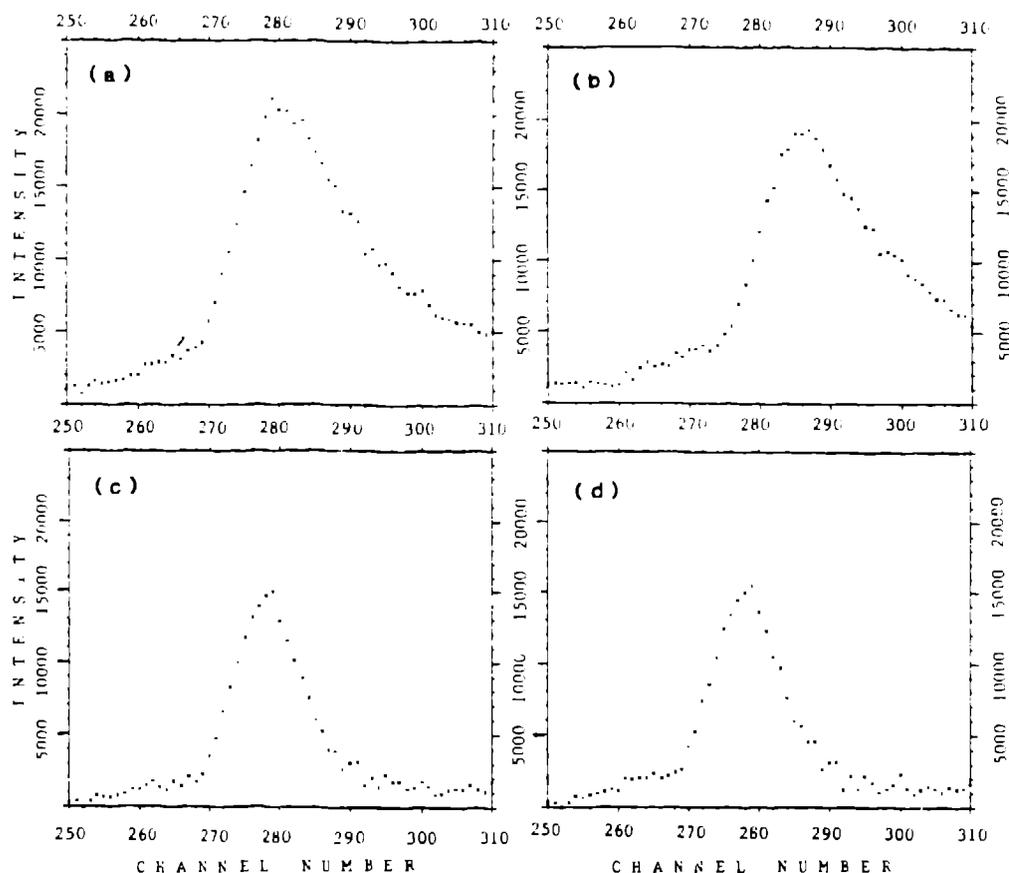


Fig. 5 Inelastic neutron scattering spectrum of the OHO bending mode in CrOOH (channel width $9.6 \mu\text{s}$)
 (a) data taken with Be filters
 (b) data taken with BeO filters
 (c) filter difference Be-BeO
 (d) mathematical filter difference

Finally, it should be pointed out that the 'window n' is a general parameter which may be chosen to match the resolution adopted to the problem.

Statistical aspects and application of this method to more recent experimental data will be given in a forthcoming paper.

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