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PRINCIPLES OF REVERSE TIME-OF-FLIGHT METHOD AND FOURIER TECHNIQUE

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ABSTRACT

A general lay-out and principle of operation of the reverse time-of-flight measurement is given, applicable particularly to studying diffraction at powdered samples and predictively also to coherent exitations at single crystal samples, both characterized by spectra made up of peaks. Conclusions on the applicability of the method at steady state sources and pulsed sources of short or long pulse duration are presented[1] .

1. Introduction

The interest in the reverse time-of-flight (RTOF) method, introduced in 1972 [1], has only recently started to grow outside of the small core of pioneers. The major impact forwards took place in 1992, when the first implementation at a pulsed neutron source, the high-resolution Fourier diffractometer HRFD was completed in Dubna and started its successful operation [2]. Powder diffraction has so far been the sole area of applications. The first successor of the early demonstrations was the mini-SFINKS diffractometer, jointly built by PNPI, Russia and VTT, Finland in Gatchina [3]. It has already been used some ten years for extensive studies on crystallography. The next installation FSS, the Fourier strain spectrometer, started its operation in Geesthacht in 1988 [4]. Currently there is an on-going project for constructing a time-of-flight Fourier diffractometer at the ET-RR-1 reactor in Cairo, Egypt [5].

Most of the published litterature on the subject deals with experiments at steady state sources. It seems, however, that best performance and widest spectrum of applications can be expected at
pulsed sources [2,6], coinciding with the trend of planned future neutron sources. There the trade between resolution and intensity has led to short pulse moderators, suffering

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from severe losses in intensity and to intensity optimized, long pulse moderators, suffering from poor inherent resolution. A spallation neutron source can normally have both type of moderators simultaneously, while a pulsed reactor lacks the short pulse option. If the source pulse is not short enough, the desired resolution is most advantageously provided by Fourier choppers, operated in the RTOF-mode. Yet, the information on the on-state of the source is very useful in screening out the "wrong" neutron histories and thereby markedly increasing the signal to noise ratio. In the RTOF-method this screening can be achieved in a very flexible manner.

Complying with the workshop character of session T3, the strategy from general to specific, rather than vice versa, was considered more appropriate for dealing the subject and arriving at specific conclutions to be assigned to different types of neutron sources. It should be realized, however, that while some of the concepts to be presented are well established already, some are at a very embryotic state lacking experimental verification or quantitative simulation. The experimental arrangement to be considered is shown in Fig. 1.

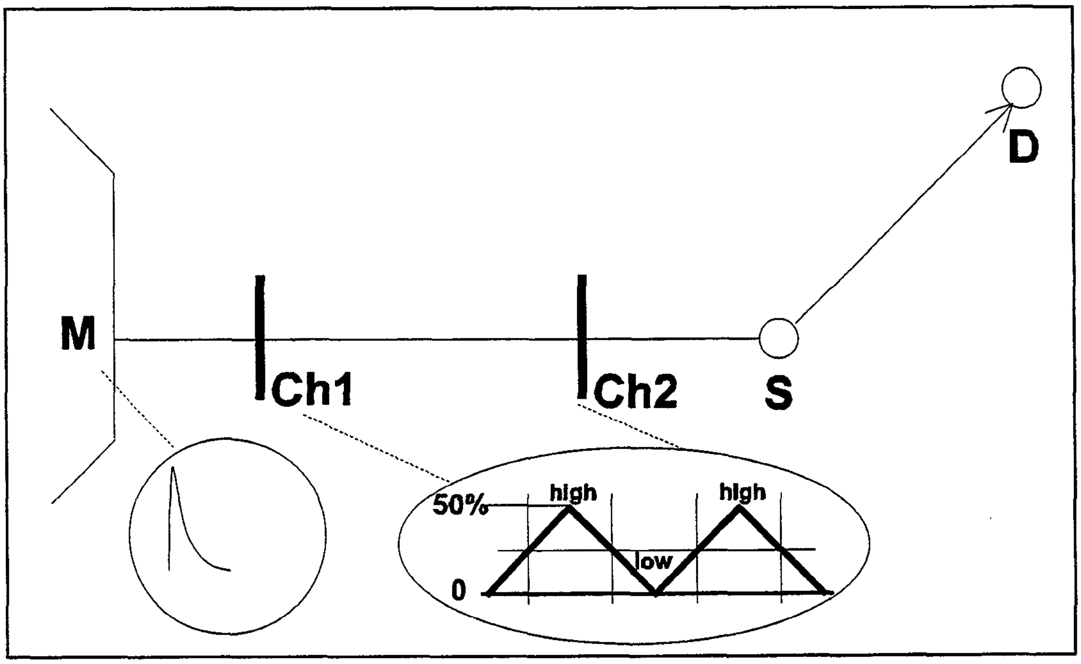

Figure 1. The schematic lay-out of the experiments under consideration. M is moderator of the source, Ch1 and Ch2 two Fourier choppers, S sample and D detector. Inserts show the time-dependence of the moderator intensity and chopper transmission.

2. RTOF-plot

The basic idea of the RTOF-method is to check for each neutron detected, whether certain postulated neutron histories have high or low probability of occurrence based on the knowledge on the actual state of the source or of the time-dependent selectors installed in the flight path. Because neutron detection occurs at the very end of its flight, checking must act on instants of the immediate past extending to the longest time-of-flight expected. Therefore the history of the states of the source and of the neutron selectors must be made available as continuously updated arrays for real time classification between the high or low probability histories. Such arrays are conveniently obtained from discrete time shift registers, provided with parallel outputs for every time channel.

The trajectories of neutrons as the travelled path length versus time-of-flight are conveniently presented in a RTOF-plot shown in Fig. 2. The figure shows the largest conceivable number of neutron selectors to be used for classification of neutron histories. Depending on the neutron source and on the purpose of the experiment the picture can be much simpler. The binary moderator delay line, labelled with M, is shown as the black horizontal ribbon, most of the time off, but shortly interrupted by the white on-states, as the burst of neutrons are emitted. The two black and white ribbons labelled with Ch1 and Ch2 are binary delay lines of two Fourier choppers. White colour indicates the high-transmission state and black

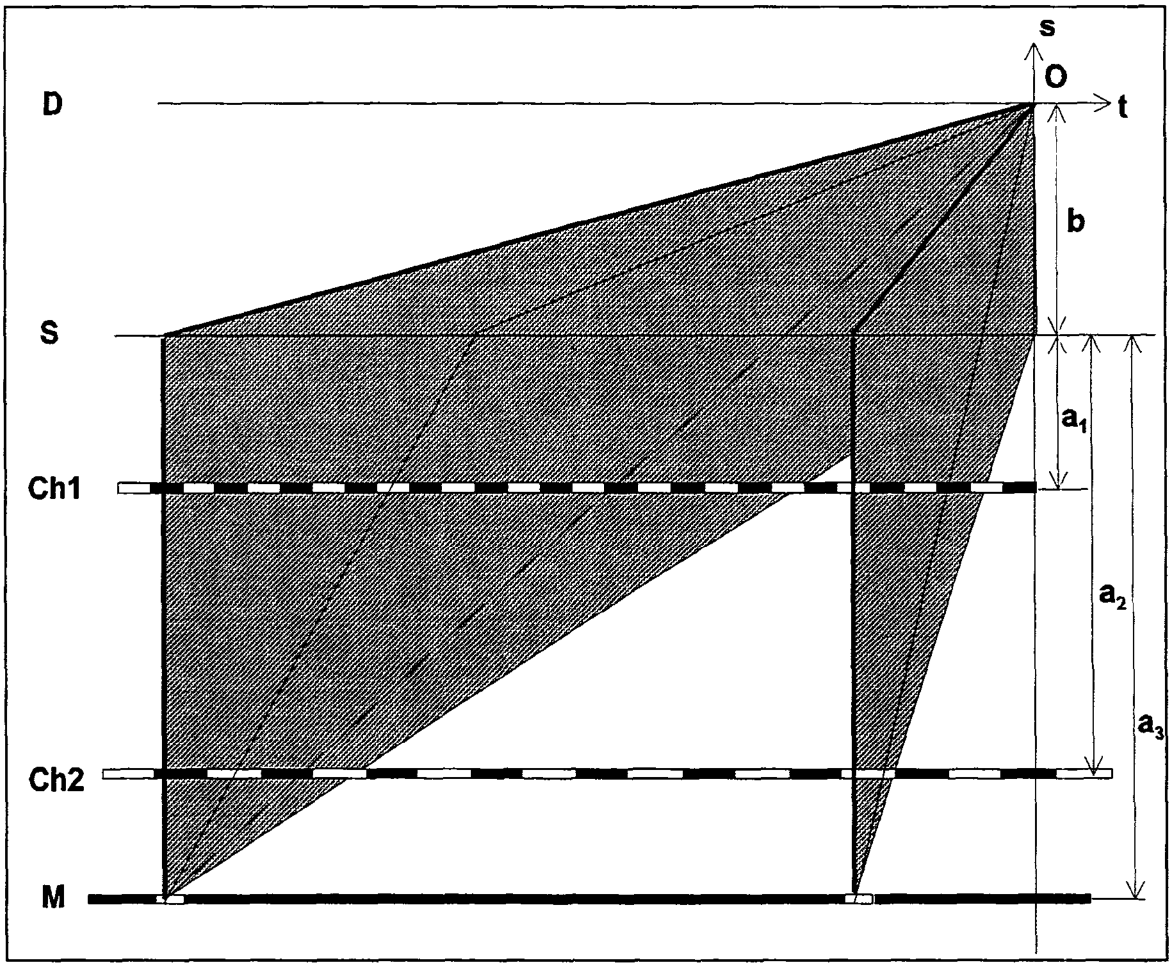

Figure 2. RTOF-plot for a pulsed source and two Fourier choppers. Horisontal lines labelled with D, S, Ch1, Ch2 and M are the locations of detector, sample, chopper1, chopper2 and moderator. Origin is fixed at the detector at the instant of neutron detection.

colour the low-transmission state of the chopper. The grey quadrangles are the allowed time-of-flight frames associated with two most recent moderator pulses. Each frame is divided by the straight, elastic scattering line into the down-scattering regime to its lef and up-scattering regime to its right. Presented in reversed time order the allowed neutron histories start from the origin as a straight line within the grey frame to any point of the sample line S and continue from there as another straight line staying within the grey frame to any white point at the moderator delay line.

Screening means that certain neutron histories are considered as impossible and are not classified or recorded at all. This applies to histories with the moderator in the off-state or, in the case of two choppers, also to histories with one of the choppers in the on-state and the other in the off-state, such as the elastic line in the most recent frame of Fig.2.

Measuring means classification of the unscreened histories in the high-probability and low-probability records. The down-scattering history in the most recent minus one frame must be classified as a high-probability event, since all selectors were in white on-state along its path. The elastic history of the same frame must be classified as the low-probability event, since both choppers were in black off-state along its path.. As the choppers are rotated according to one of the common frequency windows used in Fourier synthesis taking care that all phases of the choppers are uniformly covered without any mutual correlation, the signal or an estimate of the time-of-flight distribution is obtained by subtracting the low-probability record from the high-probability record and an estimate of its variance by adding the two together. Because the local Fourier codes of two time-of-flights outside of the resolution width are practically orthogonal, frame-overlap disappears from the signal, but is present in the variance part.

Considering now powder diffraction or one-phonon scattering at a single crystal along a chosen line in the momentum space the time-of-flight scan for one detector is allways one-dimensional. This means that one has to connect every time channel of the moderator delay line to one and only one time channel of each chopper delay line according to some simple rule chosen according to the experiment. For this adjustmet two parameters for each delay line n are available, namely the channel width $\Delta t^{(n)}$ and the predelay $d^{(n)}$. Three different scans can be accomplished namely {k_i = const.}, {k_f = const.} and { k_i / k_f = const. } with the last scan containing the elastic scan as a special case. The choice of parameters for each of the three scans are compiled in Table 1.

Table 1. Choice of the delay line parameters for the three different scans. $T^{(1,3)}{}_i$ is the incident neutron time-of-flight between components (1,3), e.t.c.

	k_i = const.	k_f = const.	$k_i / k_f = r_k$ =const.
channel width condition	$\Delta t^{(1)}=\Delta t^{(2)}=\Delta t^{(3)}$	$\frac{\Delta t^{(1)}}{a_1}=\frac{\Delta t^{(2)}}{a_2}=\frac{\Delta t^{(3)}}{a_3}$	$\frac{\Delta t^{(1)}}{a_1+r_k b}=\frac{\Delta t^{(2)}}{a_2+r_k b}=\frac{\Delta t^{(3)}}{a_3+r_k b}$
predelay condition	$d^{(1)} = d^{(3)} - T^{(1,3)}{}_i$ $d^{(2)} = d^{(3)} - T^{(1,2)}{}_i$	$d^1 = d^2 = d^3 = T^{(S,D)}{}_f$	$\frac{d^{(1)}}{a_1+r_k b}=\frac{d^{(2)}}{a_2+r_k b}=\frac{d^{(3)}}{a_3+r_k b}$

3. TOF scans in momentum space

A simple FCC lattice of Fig. 3 was chosen as a frame of reference for illustrating in the momentum space the three TOF-scans introduced above for a single detector. Scan 1 with $\{k_i = \text{const.}\}$ is not very interesting, because only one detector can measure along the same line for fixed orientation of the sample. Modifying scan 2 from $\{k_f = \text{const.}\}$ to $\{k_f\sin(2\theta) = \text{const.}\}$, brings scans 2 of all detectors, independent from θ, to coincide in the momentum space differing in their energy transfer versus momentum transfer relationship, as well known from TOF-operating spectrometers. Scan 3 is a very interesting scan directed right to the origin of the momentum space. If this scan is adjusted to go through any of the neighbouring reciprocal lattice points, only longitudinal phonons will get exited. Like before scans of detectors at different scattering angles can be adjusted to coincide in the momentum space, while their energy transfer versus momentum transfer relationship follows parabolic law. A certain scattering angle corresponds to elastic scattering allowing one to

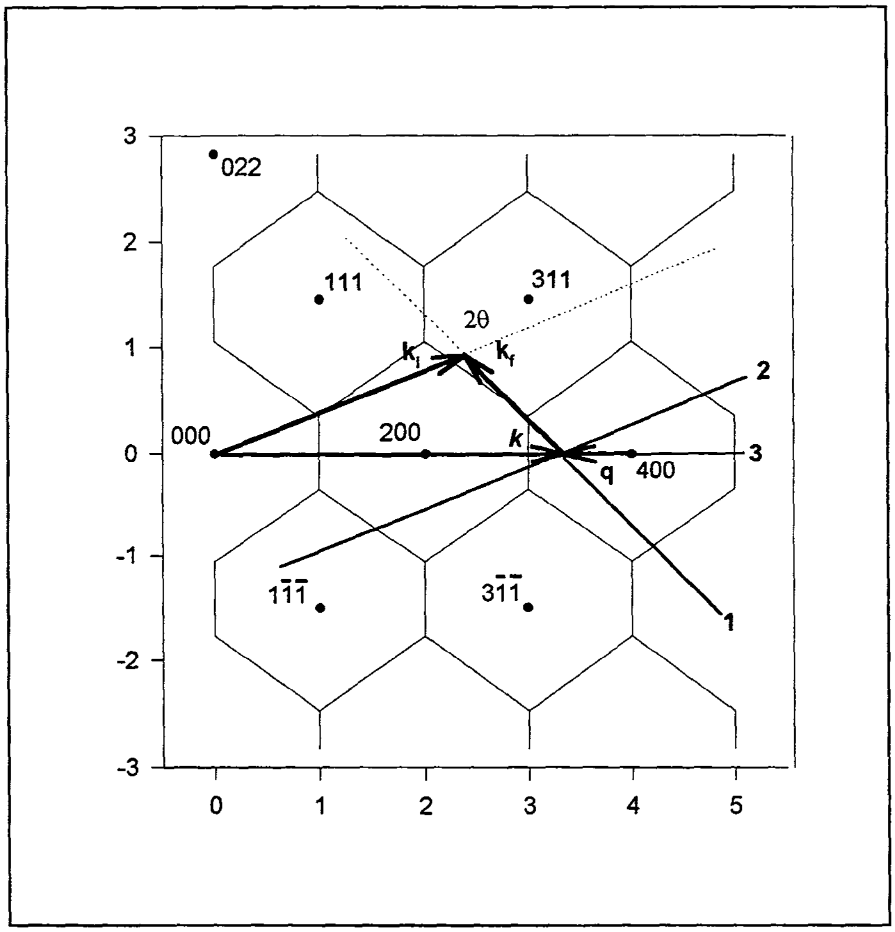

Figure 3. Reciprocal lattice plane of an FCC-crystal, perpendicular to $(01\underline{1})$-axes, with 1 labelling the $\{k_i = \text{const.}\}$-scan, line 2 the $\{k_f = \text{const.}\}$ scan and line 3 the $\{ k_i / k_f = \text{const.} \}$-scan.

accomplish a very simple diffractometer with inelasticity discrimination leading to definite signal-to-noise ratio benefit compared to the total scattering instruments, almost exclusively used to day.

3. Applicability of RTOF-method at different neutron sources

A general guide for the applicability of the RTOF-method for studies on diffraction and on phonons or magnons at different neutron sources is outlined in Table 1. The prospects of the steady state source case are limited because screening of "wrong" neutron histories implies installing an extra pulsed chopper with severe intensity losses compared to a pulsed source. On the other hand powder diffraction studies on samples of "medium complexity" seems to be very competitive compared to the other alternatives available at steady state sources.

Table 2. Applicability of RTOF-method versus neutron source type.

	STEADY STATE	LONG PULSE	SHORT PULSE
DIFFRACTION for crystallography	*Well established* Total scattering Instrument with one chopper, applicability: samples with less than about 50 parameters	*Well established* Total scattering Instrument with one chopper+ inelasticity discrimination applicability: samples upto more than 50 pameters	Only benefit could be achieving inelasticity discrimination, but not competitively with long pulse source
for residual strains	*Well established*	*Established*	No benefit
PHONONS & MAGNONS: three scans with the same instrument!	Implementation requires one Fourier chopper and one pulsed chopper. Applicability depends on comparison with TAS	Probably competitive. Low resolution with one chopper, high resolution with two choppers and modif. data acquisition	Probably competitive. High resolution with one chopper only and simple data acquisition

Considering powder diffraction at pulsed sources one should notice that due to time-focussing and large detector a whole range of wavelengths are contributing to the same Bragg reclection. Focussing condition can be met for one neutron selector at a time only. If another one is used for inelasticity discrimination, its time aperture must be made large enough, roughly matching the duration of the long pulse source, in order to avoid reduction of the effective area of the detector. Therefore intensity optimized, long pulse source seems to be the best choice for achieving inelasticity discrimination, useful especially for hydrogenous samples.

Residual strain application characterized by precise determination of small shifts of the positions of relatively few peaks fits ideally with the strengths of the RTOF-method. There high resolution and also good neutron economy are essential, because very small sample volumes are desirable. The unique possibility of synthetizing the diffraction peaks in antisymmetric shape has turned out to be very effective in precise detrermination of the peak shifts. [4]

Application of the RTOF method to phonon or magnon spectroscopy is still at a conceptual study stage. The next step needed is a simulation study for finding the latitude in the choice of the main parameters of such an instrument as well as a quantitative assessment of the expected performance.

4. References

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