CORRELATION METHODS IN TIME-OF-FLIGHT NEUTRON SPECTROMETRY

W. Glaser

A way of improving the time-of-flight neutron spectrometry technique is considered. This improvement is based on the application of correlation choppers which permit the intensity of the neutron beams of steady-state reactors to be more fully exploited. The principle of operation of these choppers is described and the experiments that show most clearly the advantage of the correlation time-of-flight methods over the conventional techniques are discussed. In conclusion, the perspectives of the future development of the correlation technique are indicated.

INTRODUCTION

There is no doubt that the prospects for pulsed reactors are very great. This is indicated by the interesting and promising experiments carried out with the fast neutron pulsed reactor at Dubna. However, there are now also several steady-state reactors with a flux of 10^{15} neutron · cm^{-2} · sec^{-1}, and it would be interesting to establish whether they could compete with the better pulsed reactors. It is not easy to compare them in all respects. The most natural way of comparing them is in experiments that use time-of-flight methods. But besides comparing, it is worthwhile considering whether the time-of-flight methods for steady-state reactors could be improved. The present paper is devoted to this question.

There are, of course, two main methods of determining energy in slow-neutron scattering experiments — the diffraction and the time-of-flight method. The latter enables one to measure many parameters simultaneously with less effort than diffraction methods, but it does have shortcomings. The continuous beam from the reactor must be split into pulses with a very large spacing so as to avoid overlapping of events that take place during the time of successive pulses. As a result, the intensity is greatly reduced. This is why diffraction methods have proved more satisfactory for stationary reactors, especially in the cases when one is interested in a small number of previously specified transfers of energy and momentum (as, for example, in phonon experiments). It is only comparatively recently that specialists in the field of neutron physics have begun to think about making a more optimal choice of the working cycle — the ratio of the useful beam time during one revolution of the chopper to the period — in time-of-flight methods. It has been found that some of the previously unused neutrons could be exploited by improving the old time-of-flight method. The idea on which the improvement is based is, in fact, new only to neutron physics; it has long been employed in communications technology and other branches of science. But a certain time had to pass before sufficiently many neutron physicists became convinced of its usefulness.

We consider communications theory and we therefore call the time-of-flight spectrum the response function. Figure 1 shows that it can be measured by essentially three methods. The first method consists of exciting the system by short pulses and measuring the response function directly. In the second, the system is excited by a sinusoidal wave and a Fourier component of the response function is measured. Finally, the system can be excited by static noise, the response function being then obtained by measuring

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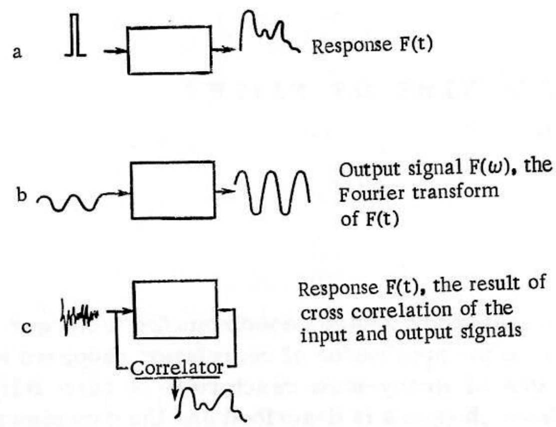

Fig. 1. Methods of determining the response function of a system: a) Direct measurement of the response function F(t) in pulse excitation; b) measurement of the Fourier transform F(ω) of F(t) in the case of sinusoidal excitation; c) measurement of F(t) by cross correlation of the input and the output signal in the case of excitation by noise.

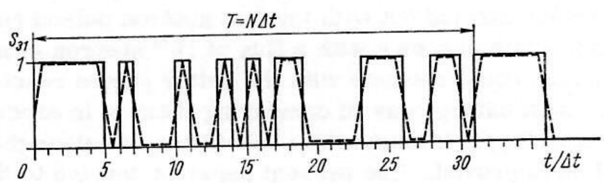

Fig. 2. Pseudorandom sequence S(t) for N = 31.

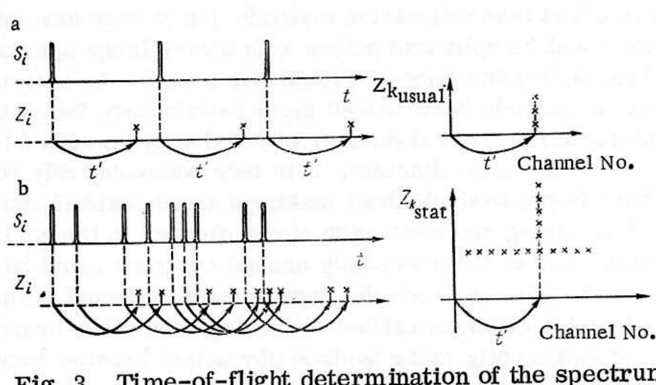

Fig. 3. Time-of-flight determination of the spectrum: a) By the usual methods; b) by statistical methods; S_i is the input signal; Z_i is the output signal; Z_k is the time-of-flight spectrum.

the cross correlation between the input and the output signals. In neutron physics one can use either the second or the third method, but I shall concentrate on the third.

In practice it is very difficult to obtain a truly statistical modulation of a neutron beam, but this is not necessary since infinite correlation times are never required. It is much more convenient to employ a modulation that is, on the one hand, random, but, on the other, periodic in time. The so-called pseudorandom binary sequences are an example; a short sequence of this kind is shown in Fig. 2. In practice, much longer cycles are used. The autocorrelation function of such a sequence is very nearly the same as the resolution function of an ordinary pulsed neutron source with exactly the same period.

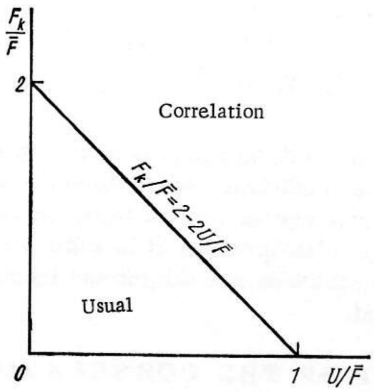

Fig. 4. Regions of applicability of the usual and the correlation methods.

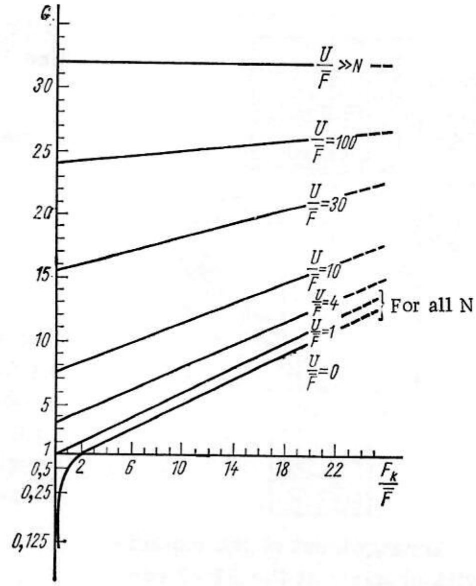

Fig. 5. Gain in time of the time-of-flight correlation method.

Rather than delving into the mathematical formalism, I would prefer to illustrate the gist of the idea for extracting the response function from the cross correlation of signals by means of Fig. 3. In the usual time-of-flight method, each input pulse gives rise to a response pulse, which, after processing in a multichannel analyzer, gives the complete spectrum.

If the sequence of input pulses is random, the finding of the cross correlation entails the finding of all possible correlations between the input and the output. The real corelations accumulate, as in an ordinary experiment; the spurious correlations give a constant background, the signal function having random properties.

This alone shows that the tremendous gain in intensity achieved by going over from a working cycle of less than 0.01 to a working cycle of 0.5 does not lead to a corresponding gain of the useful signal; spurious correlations are inevitably present. In the correlation method with working cycle 0.5, the statistical error is the same for all channels, being determined by the total number of counts. To obtain quantitative estimates of the information gain in an actual case, it is necessary to consider a definite experiment.

The absolute statistical error in the usual time-of-flight method is

$$\Delta F_k = [F_k + 2U]^{1/2},$$

where F_k is the number of events in channel k and U is the background; on the other hand, the same quantity obtained in the same measurement time in the correlation method is

$$\Delta F_k = \left[\frac{2}{N+1}\,\overline{S_i^2}\,(F_{tot}+4U)\right]^{1/2}, \quad N \gg 1,$$

where F_{tot} is the total number of counts of the response function (in the spectrum), and $\overline{S_i^2}$ is the mean square of the output signal function. The correlation method will be more advantageous if

$$\left(\frac{\Delta F_k}{F_k}\right)_{\text{pseudorand}} < \left(\frac{\Delta F_k}{F_k}\right)_{\text{usual}}$$

or

$$F_k > 2\bar{F} - 2U; \quad \bar{F} = F_{tot}/N.$$

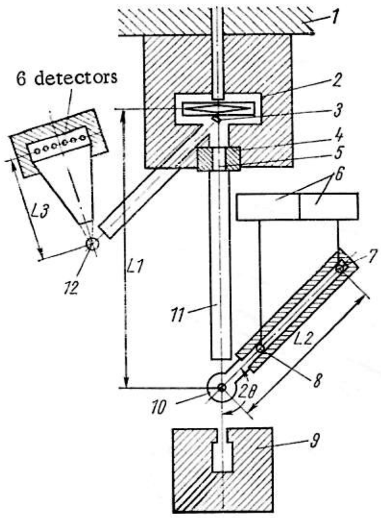

Fig. 6. Arrangement of the experiment with chopper at the FR-2 reactor; 1) reactor shield; 2) correlation chopper; 3) crystal; 4) cryostat; 5) filter; 6) time-delay analyzer; 7) detector II; 8) detector I; 9) ion trap; 10) sample holder; 11) neutron guide; 12) single crystal.

Figure 4 shows the regions in which the correlation or the ordinary method is advantageous. The boundary is the straight line $F_k/F = 2(1 - U/\overline{F})$.

In the same way one can calculate the gain in time for the same statistical accuracy:

$$G=\left(\frac{\Delta F_k}{F_k}\right)^2_{\text{usual}}\Big/\left(\frac{\Delta F_k}{F_k}\right)^2_{\text{pseudorand}}=\frac{N+1}{2}\,\frac{F_k+2U}{N\overline{F}+4U};$$

$$\overline{F}=F_{tot}/N;\quad G\approx\frac{N}{4}\quad\text{for}\quad \frac{U}{\overline{F}}>N.$$

For a cycle of length N = 127 this gain is shown in Fig. 5 for different conditions. The conditions under which the correlation methods are more advantageous are obvious: in experiment 1) with a high uncorrelated background; 2) in which resonances are sought; 3) in which resonances are sought and there is a high uncorrelated background.

1. TEST OF THE CORRELATION METHOD IN PRACTICE

We should like to complement this discussion of the principles of the correlation method by describing some practical results. Pseudorandom modulation of the neutron beam in the time-of-flight method was first carried out in independent experiments by groups in Budapest, Argonne, and Karlsruhe in 1968. In the last two cases, mechanical choppers were used. As we are best acquainted with the experiments at Karlsruhe, we shall dwell on them. Figure 6 shows the arrangement of the experiment next to the reactor, which gives a flux of 10^{13} neutron · cm^{-2} · sec^{-1}. The experiment was envisaged for phonon measurements in single crystals as well as for powder diffraction measurements, though it has so far been used for only the former.

The chopper used to give a pseudorandom pulse train is shown in Fig. 7. It consists of an aluminum disc of diameter 50 cm and it rotates at 200 m/sec at the rim. The arrangement is rather conservative and not optimal from the technical point of view, but we chose it to demonstrate the advantages of the correlation technique in different applications.

Typical results of phonon measurements in gold are shown in Fig. 8. The absorption cross section of gold is 97 b. The long vertical stroke on the right indicates the error we should have had extracting the same information by the usual time-of-flight method. The error is due principally to the uncorrelated background. It is quite clear that such an experiment could not be regarded as successful. The phonon curves of gold measured by the correlation method are shown in Fig. 9.

The results of a correlation experiment for silver are shown in Fig. 10. The absorption and the background were the same in this case as in the foregoing. The results agree well with Brockhouse's measurements with a better reactor over a longer period of time.

Figures 11 and 12 show the dispersion curves of D_2O. In this case there are no difficulties due to absorption, but the method is still advantageous, the ratio of the intensity maximum to the mean value being 4-5.

The first results in a phonon experiment being carried out currently on CsBr are shown in Fig. 13. Here again the advantage derives from the large maximum to mean ratio, the situation being actually slightly more favorable than in the case of D_2O.

The use of the apparatus has not been restricted to phonon experiments. One of the most interesting inelastic scattering experiments concerned the search for a fine structure in the para-ortho transitions in solid hydrogen due to quadrupole-quadrupole interaction of orthomolecules. Molecular rotation is hindered by this interaction, and, as earlier experiments had shown, the rotation line is broadened when the

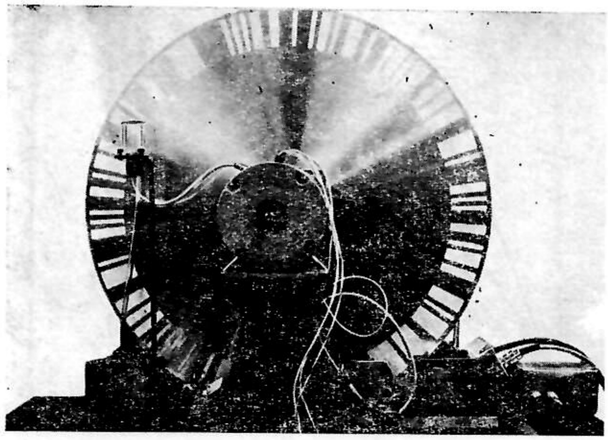

Fig. 7. Correlation chopper.

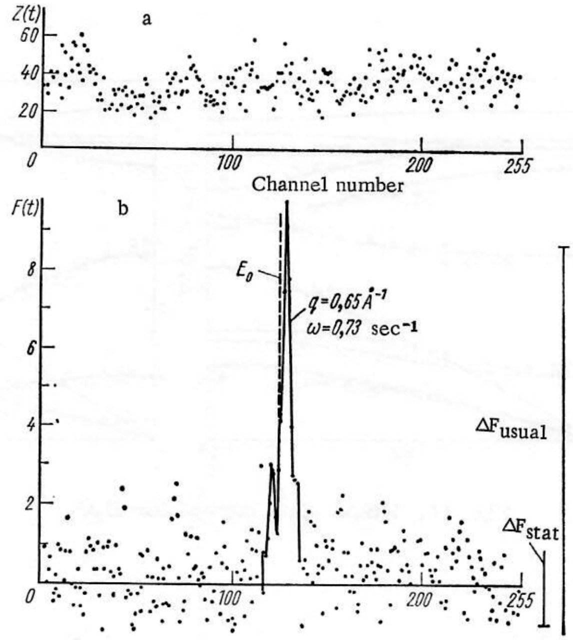

Fig. 8. Gold spectrum obtained by the time-of-flight method: a) uncorrelated pseudorandom spectrum; b) spectrum after cross correlation with the input signal.

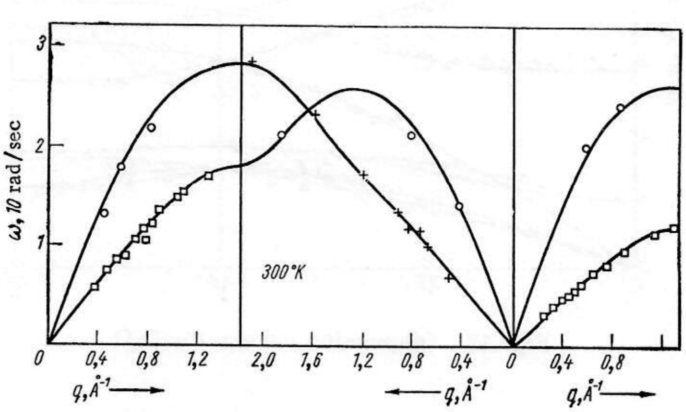

Fig. 9. Dispersion curves for gold obtained by the correlation method.

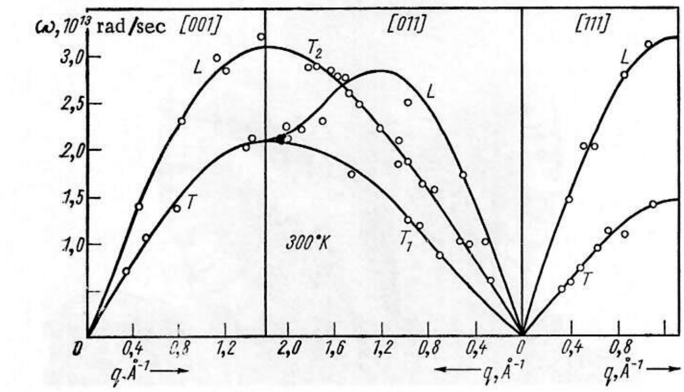

Fig. 10. Dispersion curves for silver obtained by the correlation method.

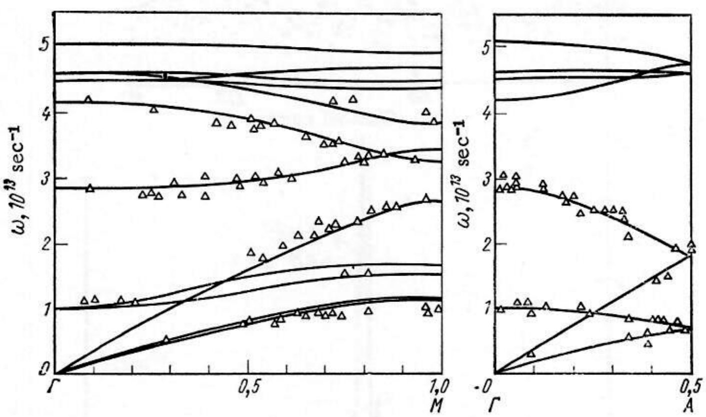

Fig. 11. Dispersion curves for D_2O.

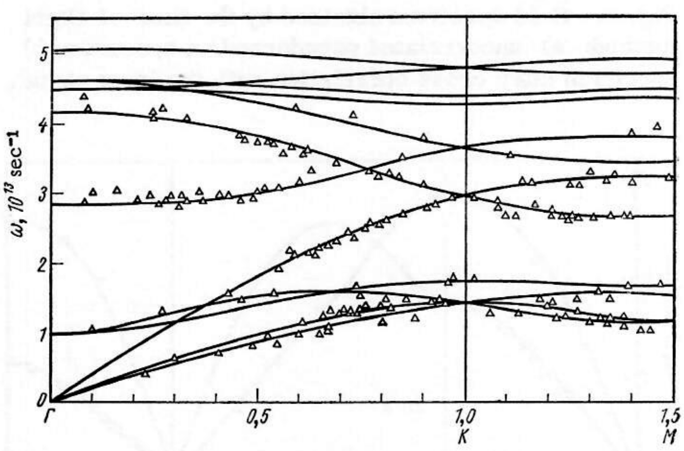

Fig. 12. Dispersion curves for D_2O.

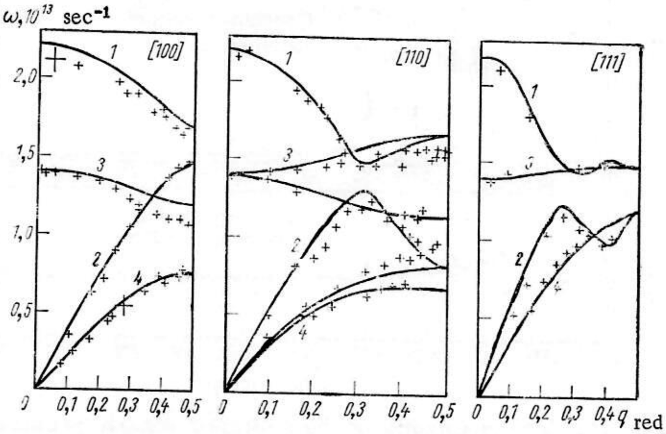

Fig. 13. Dispersion curves for CsBr at 300°K: 1) longitudinal optical branch; 2) longitudinal acoustic branch; 3) transverse acoustic branch.

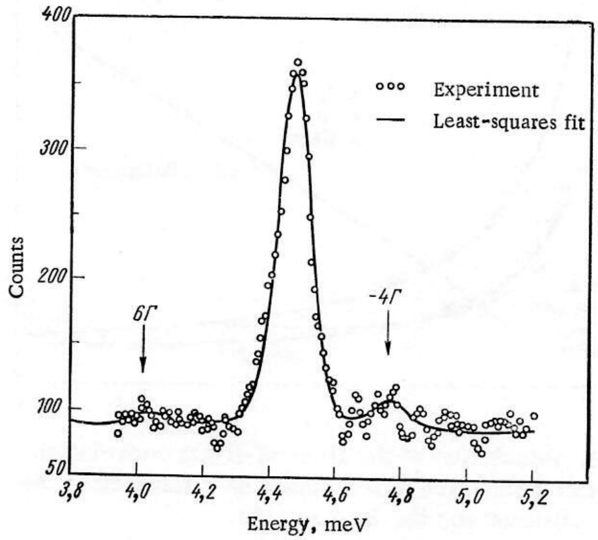

Fig. 14. Hyperfine splitting of an ortho-para transition in solid hydrogen. Two lateral peaks accompany the central peak.

orthoconcentration increases. At low orthoconcentrations, one should observe line splitting due to the interaction of pairs of orthomolecules. Therefore, a mixture containing 4% orthohydrogen was prepared for the experiment. A total of 23% of the orthomolecules were present in the form of isolated pairs. The results are shown in Fig. 14. The energy of the incident neutrons was 19.2 meV, the energy resolution 0.6%, or 100 μeV. Besides the undisplaced transition at 14.6 meV, two very small peaks were observed. They were used to determine the interaction parameter Γ, which was found to be 70 μeV.

Figure 15 shows the elastic spectrum of cadmium measured at Argonne. Figures 16 and 17 show that the correlation method is also advantageous in structural investigations.

The resolution of the part of the apparatus that works on the diffraction principle is shown in Fig. 16. This part is being currently constructed at Karlsruhe. A fragment of the $BiFeO_3$ diffraction pattern, which is similar to that observed some years ago at Dubna with a pulsed reactor, is shown in Fig. 17. It is a correlation measurement and the splitting of the [220] reflection confirms the existence of the assumed small orthorhombic deformation.

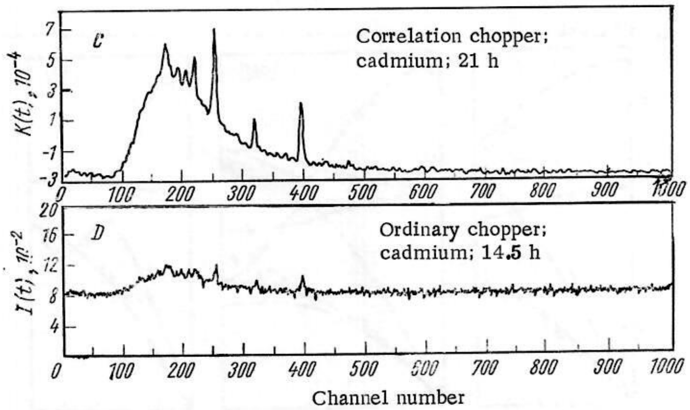

Fig. 15. Measurements of the cadmium elastic spectrum by means of a correlation and an ordinary chopper. Well resolved elastic peaks were obtained by the correlation method.

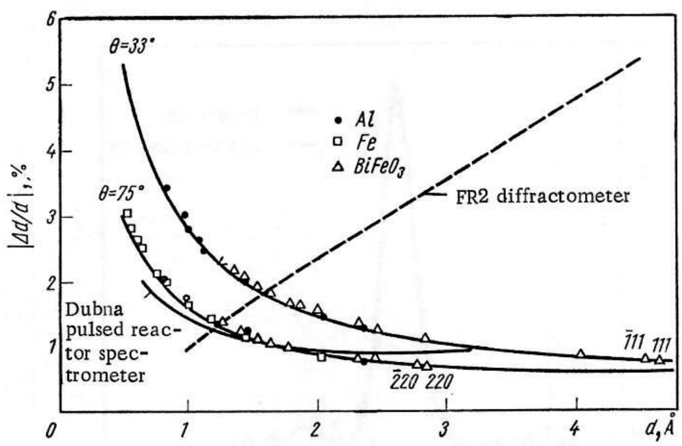

Fig. 16. Resolution of the time-of-flight correlation method in diffraction experiments as a function of the lattice constant and the Bragg angle.

2. WAYS OF IMPROVING THE CORRELATION METHOD

The advantages of the correlation method in neutron scattering experiments are obvious. However, our arrangement is not optimal in two respects.

1) The velocity of the correlation chopper at the rim – 20 m/sec – is not varied and unnecessarily restricts the beam's useful section. There would be no difficulty in increasing the rate by a factor of 2 or 3.

2) Hitherto, the working cycle has been taken equal to 0.5. However, a more detailed analysis of pseudorandom sequences has shown that 0.5 is optimal only for fairly high uncorrelated backgrounds or high ratios of the maximum to the mean value in the measured response function. In intermediate cases a working cycle less than 0.5 is optimal. For fairly flat or nonresonance response functions the working cycles can be readily calculated for different uncorrelated backgrounds. The results are as follows:

$\alpha = U/F_{tot}$	Optimal working cycle
$1/2 < \alpha$	$1/2$
$1/10 < \alpha < 1/2$	$1/3$
$1/22 < \alpha < 1/10$	$1/4$
$1/46 < \alpha < 1/22$	$1/5$
$1/82 < \alpha < 1/46$	$1/7$

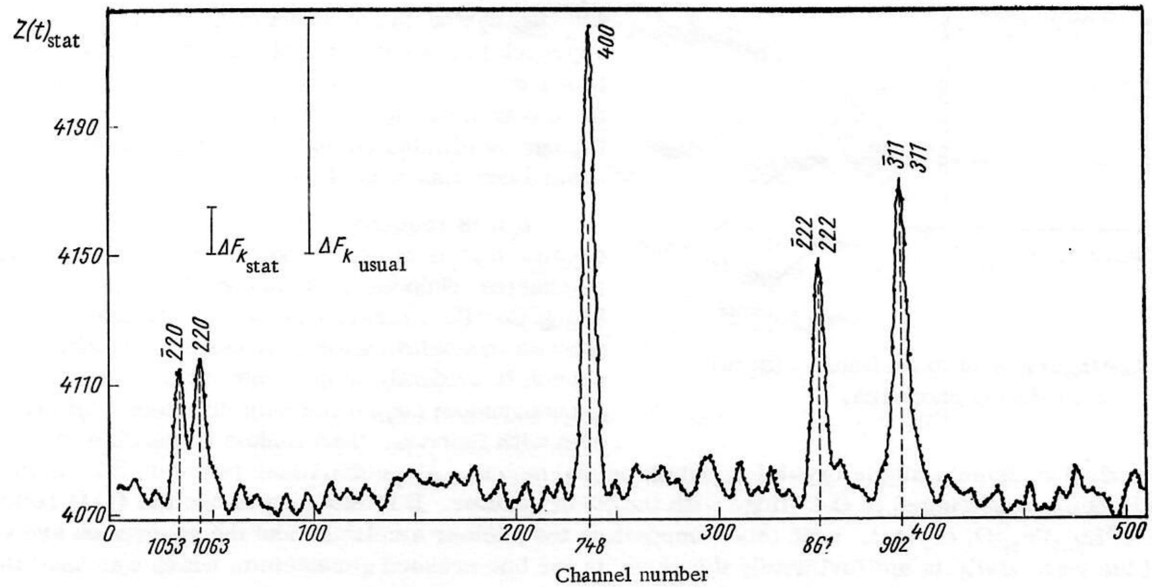

Fig. 17. Neutron diffraction pattern for $BiFeO_3$; $l = 6$ m, $\theta = 150°$, t = 27 h, G = 28.2. The measurements were made with a cooled Be filter.

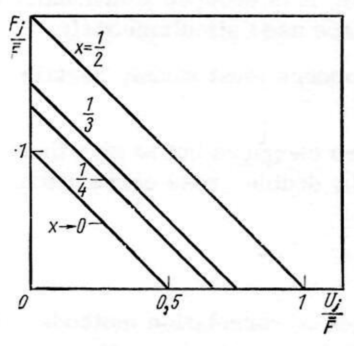

Fig. 18

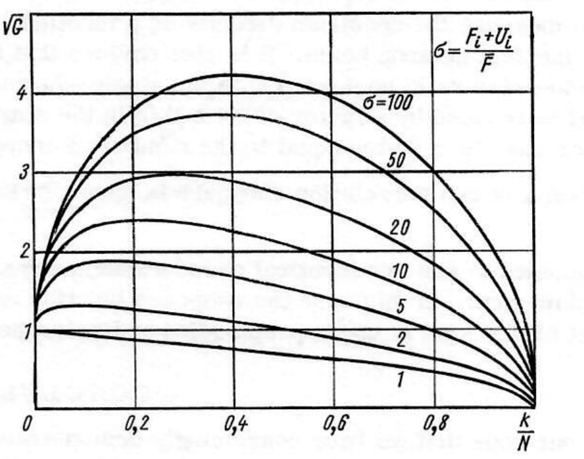

Fig. 19

Fig. 18. Region of applicability of correlation choppers with different working cycles x.

Fig. 19. Efficiency of a correlation chopper as a function of the working cycle and the ratio of the signal and the background to the mean count per channel: F_i is the magnitude of the signal; U_i that of the background; F is the mean signal at the output; $G^{1/2}$ is the coefficient of the increase of the relative accuracy; k/N is the working cycle.

More general conditions are illustrated by the following figures. The change in the region of applicability of the correlation technique when the working cycle is varied is shown in Fig. 18.

The gain in the accuracy for different working cycles and different ratios of the signal and the background to the mean signal is shown in Fig. 19 for a pseudorandom sequence consisting of 100 channels. The gain is relative to an ordinary chopper with working cycle 0.01. It can be seen from the figure that if $\sigma > 1$ there are always choppers with pseudorandom sequence that are preferable to ordinary choppers. The optimal working cycle for a fairly large class of experiments is evidently 0.2 to 0.3.

We can now draw an important conclusion: the correlation approach enables one to optimize the time-of-flight method in the planning of experiments; for example, in the solution of a problem in several

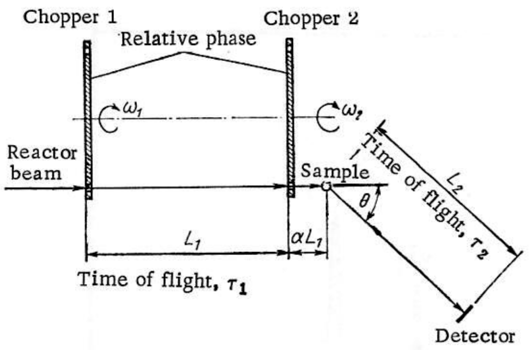

Fig. 20. Arrangement of experiment with two correlation choppers.

stages. A variety of mechanical discs with different working cycles can be prepared. However, such an approach is not very flexible and does not enable one to set up several experiments with one arrangement in an optimal manner. However, even this shortcoming can be eliminated and suggestions have been put foward how this should be done.

If it is required that the pulsed beam be monochromatic, it is better to use magnetic systems than mechanical choppers. However, the presently existing Co—Fe mirrors have a low efficiency when used as monochromators. A more promising approach is evidently to generate monochromatic pseudorandom sequences with different working cycles with ferrites. This makes it possible to use magnetic reflection from a single crystal to obtain pulsed beams. A spectrometer based on this method is currently being constructed in Oak Ridge with the HFIR reactor. It is intended to use the (111) reflection from a $^7Li_{0.5}Fe_{2.5}O_4$ crystal. With this composition the nuclear amplitude and the absorption are very small and the reflectivity is approximately the same as for hot-pressed germanium, which was used in a triaxial spectrometer. The use of this crystal makes it possible to obtain very short pulses down to 5 μsec.

One could use two and not just one chopper. In this case time-of-flight experiments would make it possible to measure the spectrum directly as a function of the primary energy contained in a polychromatic incident neutron beam. It is also obvious that there would be an even greater improvement in the signal-to-noise ratio compared with the single-chopper correlation technique. While the uncorrelated background is reduced by a factor of $(N + 1)/2$ in the single-chopper case, it is reduced additionally in the two-chopper case by a factor equal to the number of energy groups that are used simultaneously.

A system of two correlation choppers is shown in Fig. 20. The choppers must satisfy certain conditions.

We decided to use two identical pseudorandom sequences for the two choppers but to give them a phase displacement. In this case the response function can be obtained by double cross correlation. An experiment of this kind is under preparation at Karlsruhe.

CONCLUSIONS

It is our hope that we have convincingly demonstrated the advantages of correlation methods. They enable one to exploit maximally a continuous neutron beam in time-of-flight experiments. These methods have only just been introduced and much work is necessary before their full capabilities will be established.

On the basis of our own experience, we believe that this method in conjunction with high-flux denxity reactors will make it possible to measure the phonon spectra of even strongly neutron absorbing materials like Cd. Experiments concerning neutron guides could also be carried out with good resolution.

However, when pulsed reactors with a maximum intensity that exceeds that of a present-day steady-state reactor by one or two orders of magnitude with reasonable separation of the pulses have been constructed, it will be difficult for the correlation method to compete.