Method of pseudostatistical modulation in time-of-flight neutron spectroscopy

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Use of the method of pseudostatistical modulation in neutron spectroscopy is reviewed. Results are given that demonstrate the effectiveness of the method.

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INTRODUCTION

The idea of using the correlation technique for investigations in different fields of science and technology[1,2,3] and, in particular, to study scattering of slow neutrons, is not new.[4–6] There is no doubt of its promise for the solution of a number of interesting problems in solid-state physics, as is demonstrated by the wide use of the correlation method. At the present time, installations with the correlation method are used at the following scientific centers: The Argonne National Laboratory (USA); The Laue-Langevin Institute (Grenoble, France); The Center for Nuclear Physics Research (Grenoble); The Institute for Nuclear Physics (Karlsruhe, West Germany); The Technical University (Munich, West Germany); The Institute for Solid-State Physics (Jülich, West Germany); Central Research Institute for Physics (Budapest, Hungary); The Joint Institute for Nuclear Research (Dubna, USSR). The fruitfulness of the method comes out particularly clearly in the very promising applications of it with pulsed neutron sources.[7–9]

In 1972, Gläser[10] published a review article on various results obtained with correlation neutron spectroscopy, but the special nature of the selected material and the complete absence of a bibliography seriously reduced the value of this review. Therefore, we have felt it desirable to publish a new and more complete review of the method, beginning with the articulation of the idea and following up its various applications. We have tried to avoid going into details, and have considered rather the basic aspects of correlation spectroscopy; the reader can find a detailed exposition in the works listed in the bibliography.

1. BASIC IDEA OF THE CORRELATION-TECHNIQUE METHOD

To measure any neutron scattering process, one requires information about the energy and the momentum of each neutron before and after the interaction with the scatterer. Among the various ways of measuring these quantities, an important one is the time-of-flight method, which reduces the determination of the energy and the momentum to a measurement of the time-of-flight of the neutron from the source (or some equivalent position, for example, the slit of a pulse chopper) to a detector. The number of detected neutrons at time t is expressed then by means of a convolution:

$$Z(t)=\int_0^t S(t-t')\,a(t')\,dt'+b(t), \tag{1}$$

where the function $a(t')$ describes the form of the incident spectrum; $S(t)$ characterizes the physical properties of the investigated sample; $b(t)$ describes the behavior of the unmodulated background.

This situation is well known in information theory,[11] in which the problem is formulated as follows: How can one determine the characteristic function $S(t)$ of the system if a known signal $a(t)$ is fed in at the input of the system and at the output the response function $Z(t)$ is observed?

Because of the linear nature of the integral equation (1), there are many ways of determining $S(t)$; here, we shall consider three, the ones that are most widely used. The first method presupposes the sending in of a signal $a(t)\sim\delta(t)$. The second method is based on the use of a periodic input signal with successive repetition of the measurement at different frequencies and subsequent Fourier transformation of the obtained results (see, for example, Ref. 12). The present review is devoted to the third method, the correlation method. The method is based on the idea that, in principle, the function $a(t)$ may be chosen such that the autocorrelation function C_{aa} is a delta function:

$$C_{aa}(t-t'')=\int_{-\infty}^{\infty} a(t-t')\,a(t'-t'')\,dt'\sim\delta(t-t''). \tag{2}$$

It is easy to show that with this "excitation" of the system the correlation of the response function and the exciting signal has the desired form

$$\int_{-\infty}^{\infty} Z(t)\,a(t-t'')\,dt=\int_{-\infty}^{\infty} S(t')\,dt'\int_{-\infty}^{\infty} a(t-t')\,a(t-t'')\,dt$$
$$=\int_{-\infty}^{\infty} S(t')\,\delta(t'-t'')\,dt'=S(t''). \tag{3}$$

White noise has the property (2). If the mean value of the noise signal is zero, then in the procedure of correlation counting the contribution of the background $b(t)$ is appreciably reduced; in particular, if $b(t)$ is a constant, it is simply transformed to zero. In practice it is, however, impossible to make measurements with an infinitely long input signal having zero mean value, and it is necessary to replace the white noise function by something more readily available.

In fact, there does exist a finite sequence a_i consisting of zeros and units, which has the properties[13]

$$\sum_{i=0}^{N-1} a_i=m$$

and

$$\sum_{i=0}^{N-1} a_{i+j}a_{i-k}=C_{aa}(j-k)=\begin{cases} m, & \text{if}\quad j-k=0,\ \pm N,\ \pm 2N\ \ldots;\\ K & \text{in all remaining cases,}\end{cases} \tag{4a}$$

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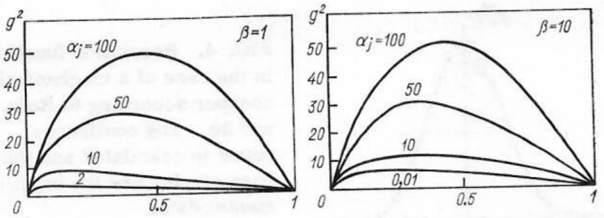


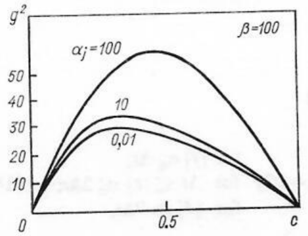


FIG. 1. Gain factor g^2 as a function of the duty cycle c for different ratios of signal to the average useful signal ($\alpha_j = \overline{S}_j/S$) and the relative background $\beta = b/\overline{S}$.

or

$$C_{aa}(j-k) = m(1-c)\,\delta(j-k) + mc. \tag{4b}$$

Here, m is the number of units in the given sequence, N is its length, and $K = m(m-1)/(N-1)$. The number $c = (m-1)/(N-1)$ characterizes the "duty cycle" of the sequence. This series, which, not very felicitously, has been called a pseudorandom sequence, is the optimal of the known binary series that approximate white noise.[14]

With this formal expression, the expression (1) is transformed to the discrete form

$$Z_n = \sum_{i=0}^{N-1} S_i a_{n-i} + b. \tag{5}$$

Here and in what follows, we shall assume for simplicity that the background does not depend on the time, i. e., on the index n.

The correlation Z_n with the sequence $\{a_i\}$ has the form

$$I_r = \frac{1}{m} \sum_{n=0}^{N-1} \frac{a_{n-r} - c}{1-c} Z_n - \frac{b}{m}. \tag{6}$$

Substituting here (5) and using the properties (4), we obtain

$$I_r = S_r + \frac{c}{1-c} \sum_i S_i - \frac{b}{1-c}. \tag{7}$$

The last expression shows that channel r contains, besides the useful signal S_r, a certain additional background line, whose value is proportional to the total useful signal. It is easy to obtain an expression for the statistical accuracy of the quantity I_i in the correlation method:

$$\operatorname{Var} I_i\,|_{(\mathrm{corr})} = m^{-1}(1-c)^{-1}\,[(1-2c)\,I_i + cNI + (1-c/m)\,b], \tag{8}$$

where $\operatorname{Var} I_i$ are the diagonal elements of the error matrix. The expression (8) enables one to compare the correlation method with the ordinary time-of-flight method. Such a comparison is usually made when one is considering a chopper of the disk type to avoid the complexities which arise unavoidably in the case of comparison with a Fermi chopper,[15] which has a different type of spatial resolution. For such a comparison, one can show that for the transmission function in the case of rectangular transitions from the position 0 to 1 and back that

$$g^2 = \frac{\operatorname{Var} I_i\,|_{(\mathrm{usual})}}{\operatorname{Var} I_i\,|_{\mathrm{corr}}} = \frac{[c\,(N-1)+1]\,(1-c)}{(1-2c) + c\,\{N + \beta\,(2 - [c\,(N-1)+1] - 1)\}\,(\alpha_i+\beta)^{-1}}, \tag{9}$$

where $\alpha_i = S_i/\overline{S}$; $\beta = b/\overline{S}$; $\overline{S} = (1/N)\sum_{i=0}^{N-1} S_i$; g^2 is called the gain factor and its magnitude characterizes the situation in which one or other of the two methods of measurement is more advantageous from the point of view of statistical accuracy.[15-20] In the simplest case ($c \sim N/2 \gg 1$ and rectangular signal) the correlation method is more effective if the following conditions, which can be readily obtained from (9), are satisfied

$$\alpha_k + \beta > 2,\; S_k/\overline{S} + b/\overline{S} > 2. \tag{10}$$

Analysis of (10) shows that the use of the correlation method is advantageous for the channels of the spectrum in which the useful signal exceeds by at least a factor two the average number of counts over the whole of the detected spectrum, and also for all channels in which the background is twice the average useful signal. Therefore, one can decide which of the two methods is the more advantageous only if one knows the background conditions and which part of the spectrum is of greatest interest.

The nonlinear dependence of the ratio (9) on the duty cycle c makes it possible to consider whether or not the process of measurement could be optimized with respect to c, depending on the background conditions. It can be shown that such optimization does indeed occur,[21-24] for which the transmitting surface must be reduced as the ratio $(S_k + b)/\overline{S}$ is reduced, i. e., its values must approach the value in the usual method, in complete agreement with the conditions (10) (Fig. 1, Table I).

2. REALIZATION EXPERIMENTS

Beam modulation is achieved in two ways: Either an absorbing substance, say Cd or Gd_2O_3, is introduced into the beam in the necessary sequence, or, in the case of polarized neutrons, their polarization is changed in accordance with a definite law.[5,25,26] In the majority of cases, the first variant is realized.[27-31] The beam is modulated by means of a rapidly rotating disk on which a pseudorandom sequence is deposited in the form of a sequence of absorbing and transmitting sections (Fig. 2). In the second case, the beam of polarized neutrons is transmitted through a spin flipper (Fig. 3). Both cases are realizable in two variants. In the first of them, the incident beam is monochromatized,[5,25-31] and in the second the incident beam is white; the energy

TABLE I.

$(S+b)/\overline{S}=\alpha+\beta=d$	Optimal duty cycle
$1/2 < d$	1/2
$1/10 < d < 1/2$	1/3
$1/22 < d < 1/10$	1/5
$1/46 < d < 1/22$	1/5
$1/82 < d < 1/46$	1/7

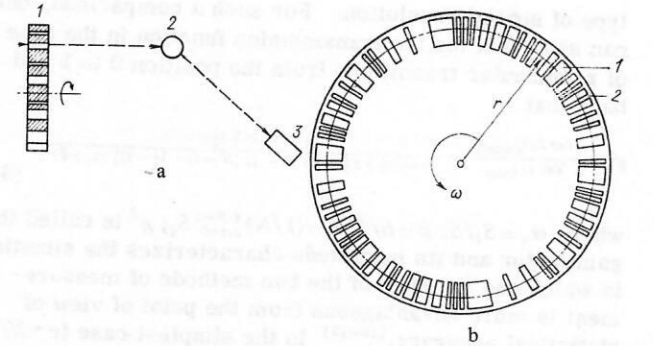


FIG. 2. Schematic arrangement of correlation spectrometer (a) with mechanical chopper, (b) on a stationary reactor. a: 1) Chopper; 2) sample; 3) detector. b: 1) Sections with covering of absorbing layer (Gd_2O_3 or Cd); 2) "transparent" sections.

of the scattered neutrons is fixed.[32] (Note, however, that all the currently operating installations use the first principle, and the inverse geometry method has so far remained only a project.)

Naturally, the transmission and resolution functions have a different form in the two methods, namely: the transmission function

$$T^R(t)=\frac{I_0}{2}(1+a(t))\begin{matrix}\nearrow \text{rectangular for "spin-flip" case}\\ \searrow \text{trapezoidal for rotating disk}\end{matrix}$$

the resolution function, which is related to the autocorrelation of the pseudorandom sequence,

$$\varphi(t)=\Phi(t)-1/N, \tag{11}$$

where

$$\Phi(t)=(1/T)\int\limits_0^T a(t')\,a(t'-t)\,dt'. \tag{12}$$

Here, T is the repetition period of the pseudorandom sequence.

For a rectangular signal,

$$\varphi^R(t)=\begin{cases}(1+1/N)(1-|t|/\Delta t) & \text{for } |t|\leqslant\Delta t;\\ 0 & \text{for } |t|>\Delta t,\end{cases} \tag{13}$$

where Δt is the time width of the smallest slit of the chopper.

The same function for a trapezoidal signal is

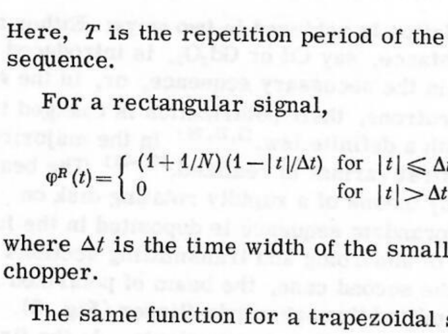


FIG. 3. Modulator of polarized neutron beam: P is the polarizer, A the analyzer, and M the control block of the spin flipper with the generator of the pseudorandom sequence.

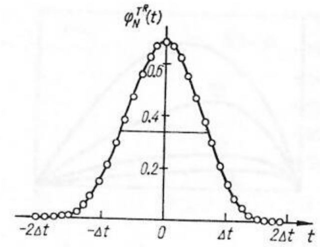


FIG. 4. Resolution function in the case of a mechanical chopper according to Refs. 5 and 30. The continuous curve is calculated and the open circles are the experimental data.

$$\varphi_N^{Tr}(t)=\begin{cases}(1+1/N)(|t|^3/2-|t|^2+2/3) & \text{for } |t|\leqslant\Delta t;\\ (1+1/N)(-|t|^3/6+|t|^2-2|t|+4/3) & \text{for } \Delta t\leqslant|t|\leqslant 2\Delta t;\\ 0 & \text{for } |t|\geqslant 2\Delta t,\end{cases} \tag{14}$$

i.e., it is nearly a Gaussian curve with width ~1.44Δt (Fig. 4).[6,30]

It can be shown that the statistical spread of the measured intensity and the form of the resolution function are related to one another.[29] To show this, we transform the expression for Var I_k to

$$\mathrm{Var}\,I_k=\frac{2}{(N+1)}\sum_{i=1}^{N}a_i^2(Z_{ik}+4b_{ik})=\frac{2}{(N+1)}\overline{a}^2(Z_{\mathrm{tot}}+4b), \tag{15}$$

where

$$Z_{\mathrm{tot}}=N\,[(N+1)\,\overline{I}/2+b],$$

and $\overline{a}^2$ is the mean square of the input signal function. For a rectangular signal $\overline{a}^2=1$, and for a trapezoidal signal $\overline{a}^2=2/3$. Therefore, Var I_k is smaller for the latter. However, this gain is offset by a worsening of the resolution [cf (13) and (14)].

With regard to the nondiagonal terms of the error matrix, i.e., cov(I_r, $I_{r'}$), it is shown in Ref. 15 that

$$\mathrm{cov}\,(I_r,\,I_{r'})=\frac{4N+1}{2\,(N-2)}\,I_0\,[S\,(r)+S\,(r')-\overline{S}]-b. \tag{16}$$

If $S(r)$ changes weakly from channel to channel, then

$$S\,(r)\sim S\,(r')\sim\overline{S}$$

and

$$\mathrm{cov}\,(I_r,\,I_{r'})\sim(1/N)\,\mathrm{Var}\,I_r, \tag{17}$$

so that the nondiagonal terms are small compared with the diagonal ones. In the opposite case, to estimate the statistical error of the parameters of the approximating curves, one must, during the mathematical evaluation of the result (for example, by the least-squares method), take into account correctly terms of the type cov(I_r, $I_{r'}$), which somewhat complicates the procedure. Equation (17) clearly demonstrates why it is necessary to take N as large as possible (to the extent made feasible by the detecting instrument). (Some problems in the calculations of correlation spectra in the case of a concrete parametrization, for example, in the presence of peaks of Gaussian profile, are considered in Ref. 33.)

One can show that it is expedient to take the width of the memory of the time analyzer an integral number of times (say, h times) smaller than the width of the ele-

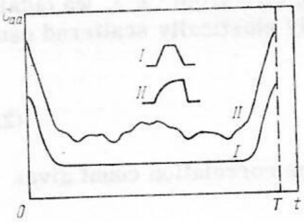


FIG. 5. Schematic profile of the autocorrelation function C_{aa} in the case of symmetric (I) and asymmetric (II) transmission functions.

mentary slit of the correlation chopper. First, if the investigated spectrum consists of sharp peaks, one needs to have about ten points on one peak in order to obtain information that is not too degraded by the worsening of the resolution resulting from the channel width of the detecting instrument. Second, there is a small additional gain in the relative statistical accuracy (rel Var I), this reaching the value 2/3 in the limit $h \to \infty$ (Ref. 15).

In the technical implementation of pseudostatistical modulation of a stationary beam it is necessary to pay particular attention to achieving a high degree of stabilization of the phase and the number of revolutions of the rotating disk. To have a good resolution, the number of rotations is taken to be ~10 000 rev/min. Therefore, a light disk of a special aluminum alloy is usually rotated in vacuum,[30] which means that numerous technical problems (such as vacuum sealing of the rotation-axis shaft) must be solved.

Another important factor is the accuracy with which the absorbing layer is deposited on the periphery of the disk. Inaccuracies (such as nonuniform thickness of the layer, ill-defined edges of the segments, and so forth) result in appreciable fluctuations in the measured spectrum,[24] and this is particularly serious if the spectrum contains strong (usually elastic) lines. One of the possible sources of fluctuations is a difference between the slopes of the leading and trailing edges of the transmission function. The autocorrelation function C_{aa} is a smooth curve if the transmission function is symmetric (case I, Fig. 5). There are fluctuations if it is asymmetric (II). The preparation of a mechanical chopper of the requisite quality is described in detail in Ref. 30.

The problem of recording and evaluating the information occupies a central position in the correlation technique. During the early stage of development of the method (in the sixties), special-purpose instruments were usually made, which, after accumulation of information by the hardware, converted the spectrum which they measured into a correlated spectrum (see, for example, Ref. 5); alternatively, the information was evaluated with a computer after the information had been transferred to its memory.[8,29] However, this procedure separates the process of measurement from the process of extracting the information, and this prevents the experimentalist from intervening in the experiment while it is proceeding. This can be overcome in an on-line experiment, which requires either fast electronics, as in the case of the reversible method,[12,34] or a computer.[35,36]

3. GENERALIZATIONS OF THE METHOD

Soon after the first reports of the successful use of the correlation technique in neutron spectroscopy, the first attempts were made to generalize the method. One direction of development was stimulated by the wish to separate the elastic and inelastic parts of the spectrum.[37,38] When the incident beam is monochromatized, an appreciable part of the spectrum is lost, and this forces one to seek new and more effective methods for applying the correlation technique. Another development was pseudostatistical modulation, which is implemented by changing the direction of the neutron polarization by means of a spin flipper, which has some particular advantages.[26,39]

Separation of the elastic and inelastic components of the spectrum. The need to filter the spectrum if it contains a strong elastic line can be seen from Eq. (8), according to which the statistical spread is basically determined by the total count. The effectiveness of the method was demonstrated by means of a computer experiment.[24]

In practice, the filtering is realized by means of two independent pseudostatistical modulators with uncorrelated pseudorandom sequences a_i and b_i (Fig. 6).[25,40] In this arrangement, the windows (transmitting slits) of one modulator control the detector, i.e., the detectors are blocked during the time of opening of the slit of this modulator with a shift corresponding to the velocity of "undesirable" neutrons.

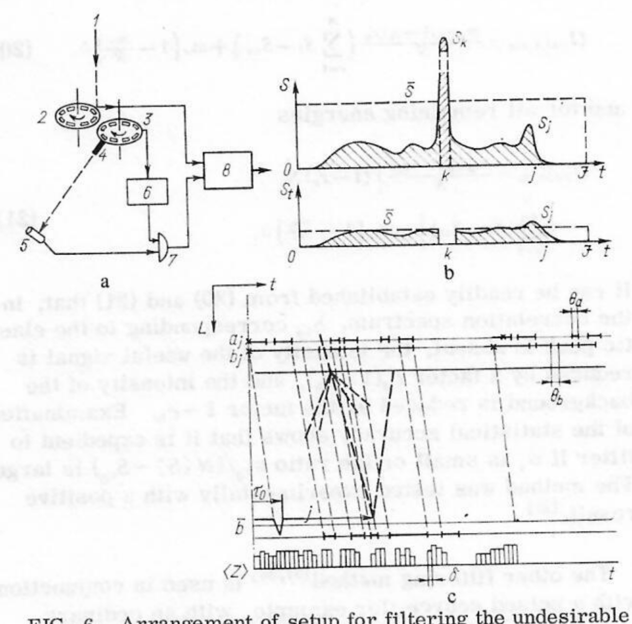


FIG. 6. Arrangement of setup for filtering the undesirable part of the spectrum by means of two modulators (a), the effect of filtering (b), time vs flight path (c). a: 1) incident beam of neutrons; 2) main modulator; 3) modulator that controls the blocking of the detectors; 4) sample; 5) detector; 6) detector blocking circuit; 7) blocking gate; 8) memory. b: $\bar{S}$ appreciably exceeds the peak S_j in channel k (upper diagram); after removal of the peak S_k (lower diagram) $\bar{S}$ is lower than S_j. c: a_j is the main modulator with width θ_a of the smallest element; b_j is the blocking modulator with width θ_b of the smallest element; δ is the channel width of the memory; $\langle Z\rangle$ is the spectrum at the detector.

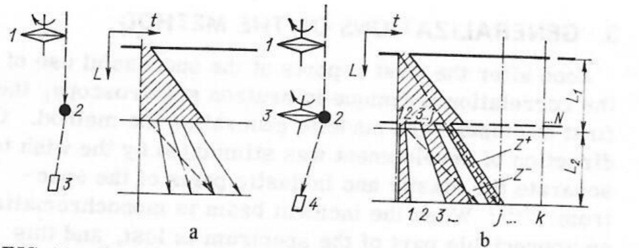


FIG. 7. Schematic arrangement and time vs flight path plot in a neutron-scattering experiment. a: 1) Chopper of ordinary type-source of a polychromatic neutron pulse whose scattering spectrum contains elastic (solid lines in the diagram on the right) and also inelastic scattering events (dashed line); 2) sample; 3) detector. b: the correlation chopper 3 placed in front of the sample 2 produces shadows (Z^-) and illuminated regions (Z^+) on the detector 4.

The measured intensity can be written in the form

$$Z_{i_1}=\sum_{i_2=1}^{N} S_{i_2} a_{i_1-i_2} b_{i_1+k-i_2}+b, \tag{18}$$

and the blocking signal has the form

$$\bar{b}_{i_1}=1-b_{i_1-\tau_0} R\left(t-t_{i_1-\tau_0}\right), \tag{19}$$

where τ_0 is the shift time; R is a function of the blocking pulse profile. If the events are detected using the pseudorandom sequence a_i, the correlation count for the channels at the forbidden energy of the neutrons leads to the result

$$(I_{Za})_{i=\tau_0}=\frac{m_a m_b (1-c_b)\, c_a}{N}\left(\sum_{i=1}^{N} S_i - S_{\tau_0}\right)+m_a\left(1-\frac{m_b}{N}\right) b, \tag{20}$$

and for all remaining energies

$$(I_{Za})_{i\neq\tau_0}=\frac{m_a m_b (1-c_b)}{N}\Big[(1-c_a) S_i - c_a \sum_{i=1}^{N}(S_i - S_{\tau_0})\Big]+m_a\left(1-\frac{m_b}{N}\right) b. \tag{21}$$

It can be readily established from (20) and (21) that, in the correlation spectrum, S_{τ_0} corresponding to the elastic peak is absent, the intensity of the useful signal is reduced by a factor $c_a(1-c_b)$, and the intensity of the background is reduced by the factor $1-c_b$. Examination of the statistical accuracy shows that it is expedient to filter if α_i is small or the ratio $s_{\tau_0}/(N\langle S\rangle - S_{\tau_0})$ is large. The method was tested experimentally with a positive result.[25]

The other filtering method[37,38] is used in conjunction with a pulsed source (for example, with an ordinary chopper placed in the beam of a stationary reactor) (Fig. 7). It can be seen in Fig. 7 that the "projection" of the chopper slits onto the detector separates the time sections which receive neutrons that have been elastically and inelastically scattered (Z^+). Only inelastically scattered neutrons hit the shadow regions Z^-. If the frequencies of the first and the second chopper do not have a common divisor (are mutually prime), it is clear that the entire incident spectrum of the neutrons will be represented with uniform weight after a time in the scattering pattern. Subtracting $\langle Z^-\rangle$ from $\langle Z^+\rangle$, we obtain the spectrum containing only elastically scattered neutrons:

$$\langle Z_j^*\rangle=\langle Z_j^+\rangle - c/1-c\,\langle Z_j^-\rangle. \tag{22}$$

The last expression after the correlation count gives

$$\langle Z_j^*\rangle=\frac{m}{N}\sum_{j'=1}^{N} S_{j'}^{(\mathrm{el})}\, S_{j-j'}+b\,\frac{m/N-c}{1-c}. \tag{23}$$

The statistical spread of the measured spectrum consists of three parts: $\sigma_1^2=(1-2c)/(1-c)\langle Z_j^*\rangle$, which is the statistical error of the elastic part of the spectrum; $\sigma_2^2=(c/1-c)\,\alpha_j\langle Z_j^*\rangle$, that of the inelastic part; and $\sigma_2^3=(1/1-c)\,\beta\langle Z_j^*\rangle$, that of the uncorrelated background.

The first application of the method and a detailed investigation of the resolution are described in Ref. 41.

Methods of obtaining two-dimensional scattering spectra. Despite the successes of the ideas described above in the correlation technique, the results so far obtained have not completely satisfied the experimentalists. To make measurements with stationary reactors, it is necessary to monochromatize the incident beam,[5,28,29] for otherwise only the elastic part of the scattering spectrum can be obtained.[33]

The first proposal for increasing the effectiveness of the method is due to Hossfeld and Amadori.[42,43] A polychromatic and pseudorandomly modulated beam impinges on a series of monochromatizing single crystals, is reflected by them, reaches the sample, and is recorded by the detector (Fig. 8a). By the choice of the distance between the monochromator and the sample one can arrange that the scattering spectra from different incident wavelengths do not overlap. In the second variant (Fig. 8b) a beam of polarized neutrons is incident on a system of magnetic single crystals whose magnetization is modulated in accordance with a pseudorandom law. Both systems are too complicated and labor-consuming for realization.

A third method, also proposed by Hossfeld and Amadori,[43] makes it possible to use a fairly large fraction of the flux of incident neutrons (more precisely, ~25%). Two pseudorandom choppers, which are arranged as shown in Fig. 8c, rotate arbitrarily. It is not necessary to introduce a phase shift between them; the imporant thing is that there should be no correlation between the two pseudorandom sequences corresponding to their modulation. (The case of nonsynchronously phased motion[28,44] is not considered here because it is of less practical value.) Every event (arrival of a neutron in the detector) can be characterized by an index k, which is related to the chopper positions as follows:

$$Z_k=\sum_{i=0}^{N_x-1}\sum_{j=0}^{N_y-1} S_{ij}\, x_{k-i-j}\, y_{k-j}+b. \tag{24}$$

The "length" of the spectrum is $N_z=N_x N_y$. The correlation count, which is made for both pseudorandom sequences, enables one to determine the scattering function

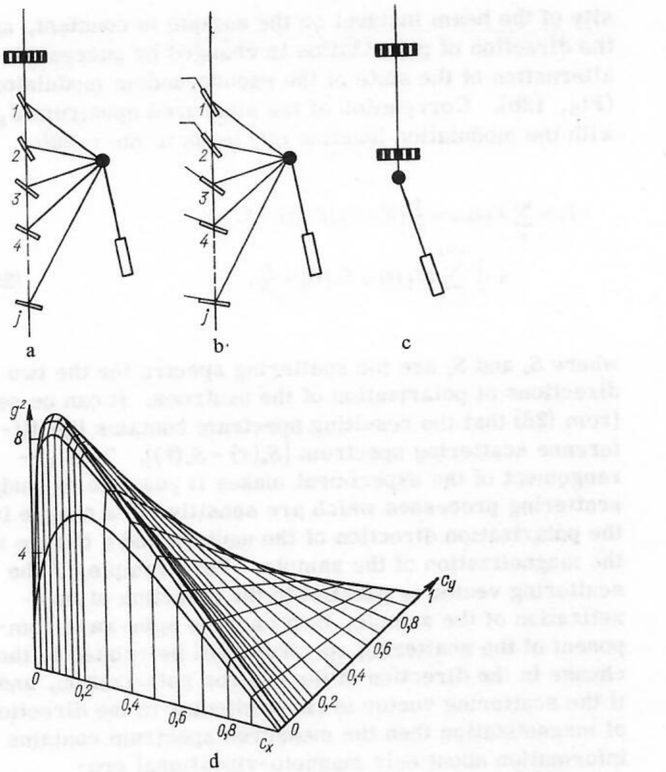


FIG. 8. Schematic arrangement with pseudostatistical chopper and monochromatizing single crystals (1, 2, ... J) (a); correlation spectrometer of polarized neutrons with magnetic single crystals (1, 2, ... J) whose magnetization is modulated pseudorandomaly (b); correlation spectrometer with two pseudorandom choppers (c) and the gain factor g^2 as a two-dimensional function of the duty cycles of the choppers (d). The g^2 surface has a clear maximum, corresponding to the optimal choice.

$$S_{uv}=\frac{1}{m_x m_y}\sum_{k=0}^{N_z-1}\left(\frac{y_{k-u-v}-c_x}{1-c_x}\right)\left(\frac{y_{k-v}-c_y}{1-c_y}\right)+\frac{b}{m_x m_y}, \qquad (25)$$

where c_x and c_y are the duty cycles of the first and the second chopper, respectively. The cumbersome expressions for Var S_{uv} and for determining the corresponding gain factor, as well as the procedure for optimizing it, can be found in Ref. 43. Figure 8d shows the surface of the gain factor above the plane of the duty cycles of the pseudostatistical choppers. The method is marred by the need to have a large memory for the detected information and the rather lengthy procedure of the correlation count, which, in its turn, determines the fate of the proposition (i. e., caused its rejection before the era of large, fast, and cheap computers).

Much more effective is the suggestion made in Refs.

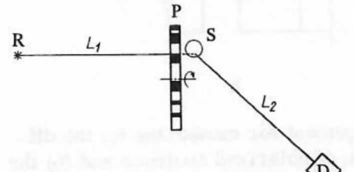


FIG. 9. Correlation spectrometer on a pulsed reactor. R is the reactor, P the pseudostatistical chopper, S the sample, and D the detector.

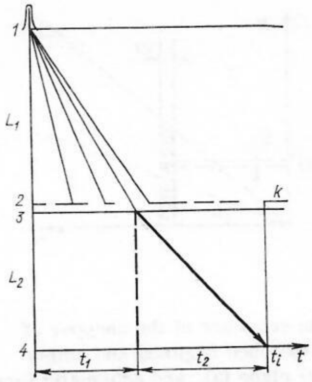


FIG. 10. Plot of flight time against flight path of correlation spectrometer on a pulsed reactor. 1) Position and time of reactor burst; 2) slits of the pseudostatistical chopper; 3) sample; 4) detector.

7 and 8 to extend considerably the scope of a pulsed neutron source by making correct use of the correlation technique (early attempts[45,46] to combine a pulsed neutron source and the correlation method did not offer the prospect of significant gain).

We place a chopper, which rotates out of phase with the periodic reactor bursts, directly in front of the sample at a distance L_1 from the reactor (Fig. 9). Suppose that neutrons recorded by a detector set up at distance L_2 from the sample are recorded in the memory with two labels: k, the position of the chopper at the time when the neutron arrives at the detector, and i, the time needed by the neutron to cover the distance $L=L_1+L_2$. Then for the scattering function (which includes as a factor the energy distribution function of the incident neutrons) and for the measured spectrum one can derive the relation

$$Z_{ik}=\sum_{j=0}^{N-1} a_{k-j}S_{ij}+b. \qquad (26)$$

As a result of the ordinary correlation count for each value of the total time of flight i we obtain

$$I_{i\tau}=\frac{1}{m}\sum_{k=0}^{N-1}\left(\frac{a_{k-\tau}-c}{1-c}\right)Z_{ik}-\frac{b}{m}. \qquad (27)$$

Simple arguments based on the diagram in Fig. 10 show that the index τ corresponds to the time-of-flight t_2 of the neutron over the second base L_2. Thus, we have at our disposal all the necessary information on each detected neutron. Indeed, suppose the scattering spectrum of the sample contains a single inelastic peak. Neutrons scattered by the sample can be detected in the cell with labels t_i and k only if the chopper was open at the time t_1 of arrival of the neutrons at the sample, i. e, if the chopper element with number $k+t_2/\Delta t_2$ corresponds to an open state (Δt_2 is equal to the time width of the shortest transmitting element of the chopper). Therefore, in the segment t_i all channels k for which the above condition is satisfied will be filled, and all the remainder will be empty. But this means that the occupation of the segment repeats the pseudorandom sequence of the chopper with a shift $t_2/\Delta t_2$, which, in its turn, is equal to the shift parameter τ in Eq. (27) (Fig. 11a).

The scattering spectrum of the neutrons lies above the t, t_2 plane, and all elastic peaks are on a straight line with slope L_2/L. Above this straight line, we have the events with loss of energy; below it, those in which the

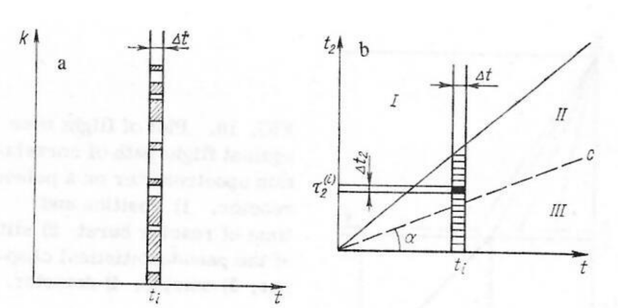


FIG. 11. Shifted pseudorandom sequence of the chopper of segment i in the form of filled (hatched regions) and empty cells of the memory on the (k, t) plane (a), and schematic form of the spectrum of scattered neutrons on the (t_2, t) plane (b). Δt is the channel width; channel $\tau_2^{(i)}$ contains a sharp peak; region I does not contain physical information; region II contains scattering events in which the neutrons lose energy; region III, where they gain energy; c is the locus $\tan\alpha = L_2/(L_1 + L_2)$ of elastically scattered neutrons.

neutron acquires energy. It is obvious that only the points of the (t, t_2) plane for which $t_2 \leqslant t$ correspond to real events (Fig. 11b). Because of this, the scattering spectrum is "squeezed" into an eighth part of the plane. The spectrum itself consists of M uncorrelated segments, where M is the number of time channels. Correlation occurs only within the segments. Since, in this case, the peaks within one segment (usually, one elastic peak accompanied by inelastic peaks) are statistically dependent, it appears reasonable to define the resolution of the method in a somewhat unusual manner,[49] i.e., as the ratio of the width of the peak Γ (say, the widest) to the distance δt of the given peak from the elastic peak in the same segment. One can express δt in terms of instrumental parameters and the energies of the incident and the scattered neutron:

$$\delta t = \sqrt{c}\,(L_1 L_2/L)\,(1/\sqrt{E_1} - 1/\sqrt{E_0}). \tag{28}$$

It is easy to show that the optimal value of the resolution is obtained for $L_1 = L_2$ and $L_1 + L_2 = L \to \infty$.

We said earlier that the chopper must not rotate in phase with the reactor bursts. This is because the chopper must serve equally as a pseudostatistical modulator for all neutrons with different wavelengths. This condition can be satisfied by the choice of the ratio of the periods of rotation of the chopper and the neutron bursts of the reactor (the periods must not have a common divisor), but if the frequency of the reactor bursts is unstable, the phase between a reactor burst and the start pulse of the chopper will vary in a random manner. In this case, if the neutron beam has a high intensity, as, for example, from an IBR-2 reactor, the statistical accuracy is basically determined by the statistical spread of the phase which is specified by the number of revolutions of the chopper.[49]

Methods that exploit features of polarized beams of neutrons. If the pseudostatistical modulation is realized by changing the direction of polarization of the neutrons by means of a spin flipper, the sample can take the position of an analyzer crystal (Fig. 12a). The total intensity of the beam incident on the sample is constant, and the direction of polarization is changed by successive alternation of the state of the pseudorandom modulator (Fig. 12b). Correlation of the measured spectrum Z_k with the modulation function $a(t)$ leads to the result

$$I_\tau = \sum_k Z_k a_{k-\tau} = \frac{1}{2}(N+1)\,[S_+(\tau) - S_-(\tau)] + \frac{1}{2}\sum_{\tau=0}^{N-1}[S_+(\tau) + S_-(\tau)] + \frac{b}{N}, \tag{29}$$

where S_+ and S_- are the scattering spectra for the two directions of polarization of the neutrons. It can be seen from (28) that the resulting spectrum contains the difference scattering spectrum $[S_+(\tau) - S_-(\tau)]$. This arrangement of the experiment makes it possible to study scattering processes which are sensitive to a change in the polarization direction of the neutron and a change in the magnetization of the sample. For example, if the scattering vector is parallel to the direction of magnetization of the sample, then only the spin-wave component of the scattering spectrum will be related to the change in the direction of the neutron polarization, and if the scattering vector is perpendicular to the direction of magnetization then the measured spectrum contains information about only magneto-vibrational processes.[39]

Naturally, the proposed method can also be used effectively to measure elastic scattering, and in some special cases, if the correlation spectrometer and the triaxial spectrometer have equal resolutions, the aperture of the former may exceed the aperture of the latter by two orders of magnitude.[26]

4. RESULTS OF EXPERIMENTS MADE BY MEANS OF THE CORRELATION METHOD

In the preceding sections, we have considered the main advantages of the method of correlation spectrometry. Numerous examples that demonstrate the advantages can be given.

In the case of strongly absorbing materials, the uncorrelated background reaches huge values, with the

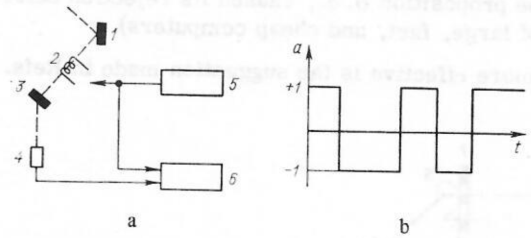


FIG. 12. Schematic arrangement for measuring (a) the difference scattering spectrum of polarized neutrons and (b) the direction of polarization of the modulated neutron beam as a function of the time t: 1) polarizer; 2) spin flipper; 3) sample; 4) detector; 5) generator of the pseudorandom modulator; 6) memory and correlator.

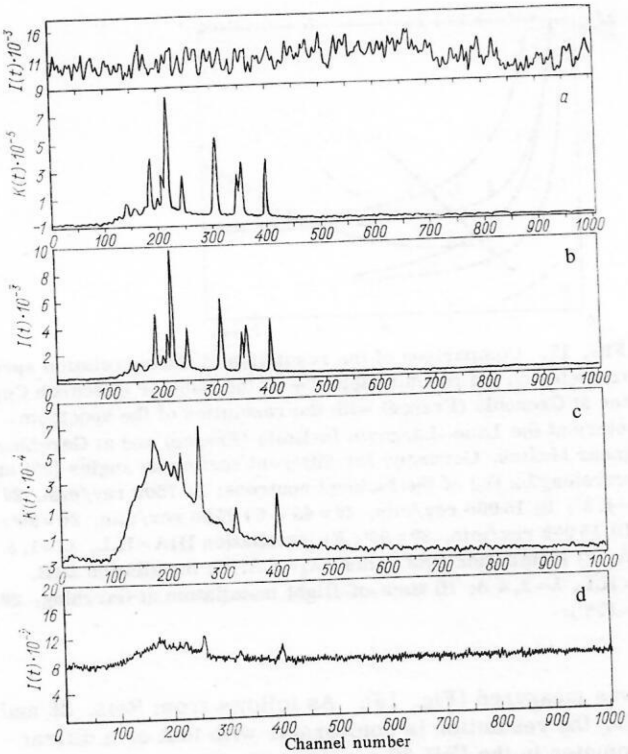


FIG. 13. Neutron-diffraction spectrum. a) Scattering on polycrystalline aluminum (correlation method, 14 h of measurement; b) on polycrystalline aluminum (ordinary time-of-flight method, 10 h of measurement); c) on polycrystalline cadmium (correlation method, 21 h of measurement); d) on polycrystalline cadmium (ordinary time-of-flight method, 14.5 h of measurement).

result that in the usual time-of-flight method the statistical spread of the experimental points due mainly to the background swamps the expected effect. This can be well seen in the scattering spectrum of polycrystalline cadmium (Fig. 13d). The same spectrum but obtained by correlation spectrometry (Fig. 13c) contains a number of sharp diffraction peaks, which can be clearly separated from the general background. However, for aluminum there is no appreciable difference between the diffraction spectra measured by the two methods; this is because of the low background, since in accordance with the condition (10) the correlation method has the advantage only at points on the upper half of the sharp peaks (Figs. 13a and 13b).[27]

A similar situation (sharp peaks in the case of a large uncorrelated background) arises when one is investigating the structure of crystalline materials under high pressure or at high temperatures. As a rule, the experimental conditions (small volume of the sample and thick walls of the high pressure chamber or the furnace) reduce the effect considerably. Here again the correlation method can be of assistance. In Ref. 31, an arrangement is described that is intended for experiments under high pressure and at high temperatures. Preliminary results (determination of the temperature dependence of the lattice parameters of aluminum oxide at high temperatures) confirm what we have said.

Work in this direction has only just begun, and there are great possibilities for its further development.

This method can also be used to investigate changes in the dynamical properties (phonon and magnon spectra) of materials under pressure. So far, dispersion curves of phonons have been investigated at normal pressure for a number of strongly absorbing materials such as gold and silver (Fig. 14).[10,28,47] According to Ref. 10, the statistical spread in the case of the equivalent ordinary time-of-flight method would be comparable with the height of the phonon peaks; for the correlation measurement it is five times less. The correlation method is also advantageous in the measurement of the phonon spectrum of frozen D_2O and cesium bromide, for which the ratio of the peak to the average background is about 4 to 5 (Ref. 10).

The importance of investigating the dynamical behavior of liquid ^{3}He by means of neutron scattering in order to obtain a better understanding of the properties of quantum liquids was recognized long ago. However, because the ratio of the cross section of coherent scattering to the absorption cross section is small ($\sim 10^{-3}$), it was difficult to make the experiments.

Measurements made by means of the correlation technique on the CP-5 reactor at the Argonne National Laboratory (it has a moderate power) at 0.015 °K led to the observation of zero sound and spin-fluctuation scattering in the continuum with excitation of particle–hole pairs in liquid ^{3}He. Figure 15 shows the neutron-scattering spectrum corresponding to excitation of particle-hole pairs.[15]

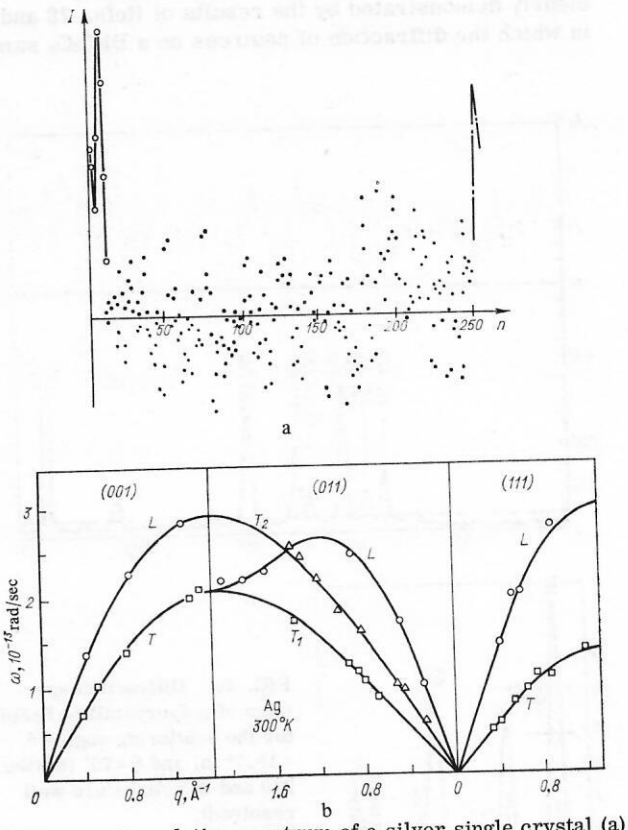


FIG. 14. Correlation spectrum of a silver single crystal (a) and dispersion curves of the silver phonon spectrum obtained by means of the correlation technique (b). The sharp peaks (a) correspond to phonons.

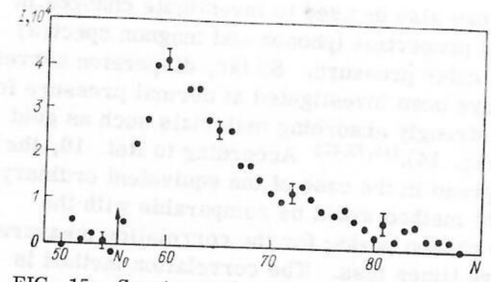
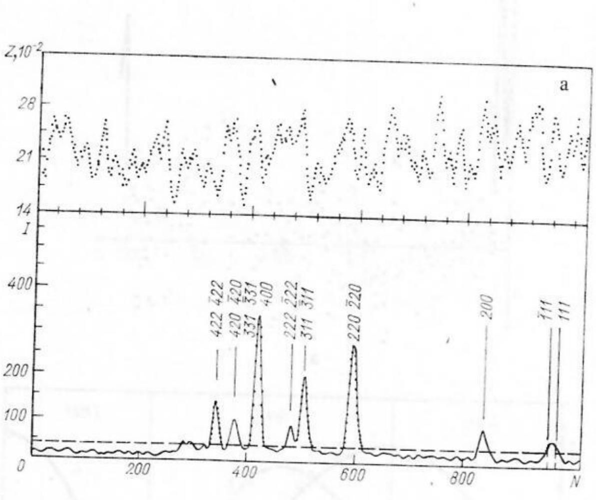


FIG. 15. Spectrum of neutron scattering on liquid ^{3}He: $I = I^3_{\text{He}+\text{container}} - I_{\text{container}}$; N is the channel number; 20 days of measurement; N_0 is the channel in which the elastically scattered neutrons fall.

It is a remarkable property of the correlation technique that the resolution can be improved by a worthwhile amount without decreasing the intensity of the incident neutron flux. The improvement in the resolution is achieved by increasing the rate of succession of the zeros and units of the modulating device with a simultaneous lengthening of the pseudorandom sequence. This last requirement is more readily satisfied for magnetic modulation of the incident beam since in the case of a mechanical chopper it is necessary to make a new disk with a modified sequence, whereas in the case of modulation of a polarized beam it is sufficient to change the switching program. (In fact, the spin-flip method is in all respects more flexible. Its only flaw is the low intensity of beams of polarized neutrons.)

The possibilities of a diffraction experiment are clearly demonstrated by the results of Refs. 28 and 29, in which the diffraction of neutrons on a $BiFeO_3$ sample



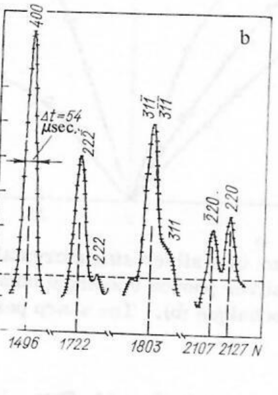


FIG. 16. Diffraction spectrum of polycrystalline $BiFeO_3$ for the scattering angles $\theta = 15.7°$ (a) and $\theta = 75°$ (b) (the 220 and $\bar{2}20$ peaks are well resolved).

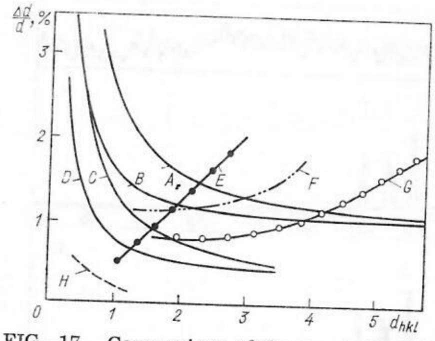


FIG. 17. Comparison of the resolution of the correlation spectrometer on the reactor Melusine at the Nuclear Research Center at Grenoble (France) with the resolution of the spectrometers at the Laue–Langevin Institute (France) and at Garching (near Munich, Germany) for different scattering angles (2θ) and wavelengths (λ_0) of the incident neutrons: A) 7500 rev/min, $2\theta = 4.5°$; B) 15 000 rev/min, $2\theta = 45°$; C) 7500 rev/min, $2\theta = 90°$; D) 15 000 rev/min, $2\theta = 90°$; E) installation D1A = ILL, $\lambda_0 = 1.5$ Å; F) installation DN3 = Siloe, $\lambda_0 = 1$ Å; G) installation D1B = ILL, $\lambda = 2.4$ Å; H) time-of-flight installation at Garching, $2\theta = 178°$.

was measured (Fig. 16). As follows from Refs. 28 and 29, the resolution is comparable with that of a diffractometer in the IBR-30 reactor even for a moderate number of revolutions and actually exceeds it for large values of the interplanar distances.

One of the main properties of the correlation method is that the resolution can be improved without loss of intensity by a simultaneous increase in the number of revolutions and the length of the pseudostatistical sequence. For example, Buevoz and Roult[31] mention a spectrometer whose chopper has 511 slits and whose error of determination of $d_{hkl} \sim 3$ Å attains $\Delta d/d \sim 0.35\%$ at 15 000 revolutions per minute. Figure 17 shows the resolution of the spectrometer. For comparison, we also show the characteristics of some modern diffractometers.

Successful testing of filtering methods is reported in Ref. 25. Figure 18 shows a spectrum containing a large elastic peak and the same spectrum after removal of the peak.

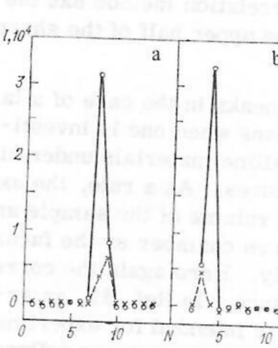


FIG. 18. Demonstration of the method of filtering a strong peak by means of two correlation choppers.[25] The intensity of the strong elastic peak is reduced to 3.2% at width $\Delta t_1 = \Delta t_2 = 50$ μsec of the windows of the two choppers (a), while for $\Delta t_1 = \Delta t_2 = 500$ μsec the intensity of the peak is reduced to 0.2% (b); the open circles represent the spectrum without filtering; the crosses the spectrum after filtering.

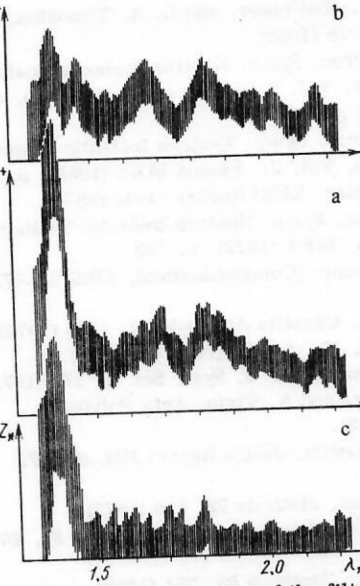


FIG. 19. Demonstration of the filtering method in the case of neutron scattering on α quartz.

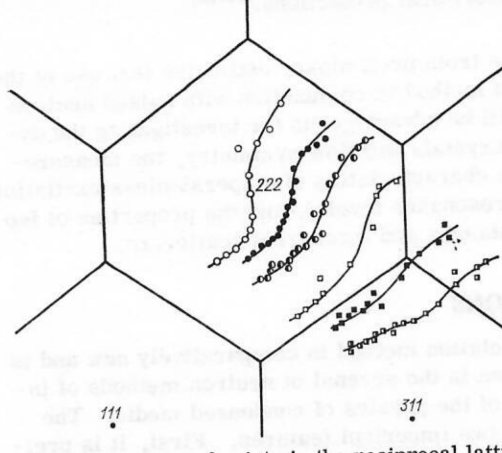


FIG. 21. Locus of points in the reciprocal lattice of aluminum near the angle (222) for measured phonons for different orientations of an aluminum single crystal: $\theta = 60°$; φ is the angle of rotation of the aluminum single crystal about the (110) axis. A minus sign in front of φ corresponds to counterclockwise rotation; the open circles, $\varphi = 6°$; the black circles, $\varphi = 10°$; the half moons, $\varphi = 15°$; the open squares, $\varphi = -6°$; the black squares, $\varphi = -10°$; the half-black squares, $\varphi = -15°$.

Pellionisz's method[37] was realized by Schneider.[41] For this, the time-of-flight installation on the reactor at Munich,[50] which consists of a long reflecting neutron guide and choppers, was slightly modified. The pseudostatistical chopper was placed immediately in front of the sample, and the neutrons were collected in two different time analyzers. (Note that the setup did not ensure optimal conditions for the experiment.) As a sample, α quartz was chosen. The measured spectra Z^+ and Z^- (Fig. 19) contain three and two peaks, respectively. The inelastic peaks are removed from the difference spectrum Z_*, and the elastic peak with an admixture of the diffuse inelastic background of very low energy is retained.

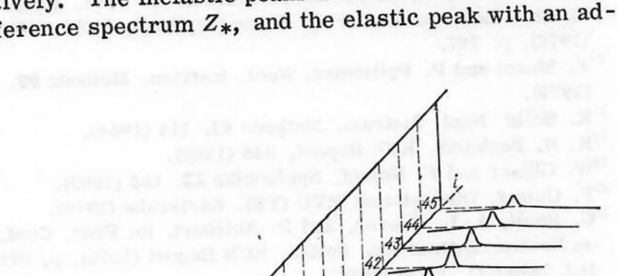


FIG. 20. Two-dimensional spectrum of neutrons scattered on plexiglass obtained by means of correlation spectrometry on a pulsed reactor at Dubna[8]: i is the channel number of the total time-of-flight (channel width, 256 μsec); t_2 is the channel number of the time-of-flight on the second flight path (channel width, 380 μsec); I_{i,t_2} is the scattering spectrum after the correlation count.

Unfortunately, some ideas for improving the correlation technique,[24,32,39,43] including a spectrometer with two correlation choppers, etc., have not been realized. In some cases, only results of computer simulation are known.[24]

One of the most promising methods—two-dimensional analysis of the neutron-scattering spectrum by means of correlation spectrometry on a pulsed reactor—is approaching completion. In 1972, a report was published[7,8] on testing of the method in the case of measurement of elastic incoherent scattering (Fig. 20).

A number of measurements of the phonon spectrum of aluminum have now been made with a moderate resolution.[36] In Fig. 21, we show the locus of points in the reciprocal lattice for measured phonons at different orientations of the single crystal. Among the large number of phonons, one can readily select phonons whose momentum vector coincides with a symmetry direction of the crystal, which makes it possible to compare the obtained data with the results of investigations with a triaxial spectrometer (Fig. 22).[48]

Study of the behavior of the resolution as a function of

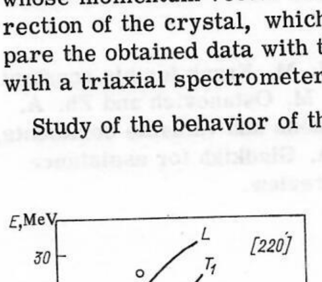


FIG. 22. Dispersion curves of the phonon spectrum of aluminum in the symmetric direction (111): the curves are from Ref. 48 and the points were obtained with the correlation spectrometer at Dubna on the IBR-30 reactor.

the spectrometer parameters reveals good agreement with the theoretical predictions.[26,49]

It follows from preliminary estimates that use of the correlation method in conjunction with pulsed neutron sources will be advantageous for investigating the dynamics of crystals with low symmetry, the measurement of the characteristics of dispersionless excitations (local and resonance levels), and the properties of isotropic substances and incoherent scatterers.

CONCLUSIONS

The correlation method is comparatively new and is a new weapon in the arsenal of neutron methods of investigation of the physics of condensed media. The method has two important features. First, it is preferable when there is high uncorrelated background, or if the physical information in the spectrum comes in the form of sharp peaks. The second feature is, in our opinion, the possibility of making a two-dimensional measurement of the neutron-scattering spectrum by means of either two pseudostatistical choppers on stationary reactors, or one in the case of pulsed sources.

In fact, these two features determine when the method should be used. The ratio of the background to the effect is large if the sample contains nuclei which absorb neutrons strongly, if the sample is small, or if under the experimental conditions the sample is surrounded by a large amount of material of a different kind (for example, in measurements at high pressures and high temperatures).

The two-dimensional methods significantly increase the efficiency of exploitation of the neutron flux of any source. However, the expected gain must be paid for by much more complicated apparatus.

Turning to the results achieved in experiments, it must be said that, despite the wide use of the correlation method, there are yet comparatively few results, which is also due to the complexity of the instruments. However, this difficulty can be overcome because of the rapid development of complicated electronic microcircuits. And the increasing number of intense neutron sources provides a real possibility for making highly effective installations.

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