### Moderation and diffusion of neutrons. Nuclear chain reactions

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A brief review is given of the investigations into neutron transport in various media and systems made by F. L. Shapiro and his collaborators. Some trends in their subsequent development, which were important both purely scientifically and for the solution of various applied problems, are indicated. The main objects of these investigations were uranium-graphite multiplying systems and the moderation and diffusion of neutrons from a pulsed source in pure moderators. © 1995 American Institute of Physics.

> Dedicated to the memory of Fedor L'vovich Shapiro by a grateful student.

#### INTRODUCTION

Investigations into neutron transport in matter (moderation, diffusion, and chain reactions in multiplying systems) make up a significant proportion of the scientific legacy of Fedor L'vovich Shapiro. Such investigations developed on a broad front in the forties in the USSR in connection with the requirements of the physics and technology of nuclear reactors that arose at that time. In particular, uranium-graphite multiplying systems were studied at the I. M. Frank Laboratory of the P. N. Lebedev Physics Institute, where Fedor L'vovich became a graduate student in 1945. Indeed, it was to investigations in this field that the first large series of his studies was devoted; subsequently, some of them formed the basis of his candidate's dissertation. 1) In the fifties, when the development of pulsed techniques had opened up the possibility of studying nonstationary neutron transport, Shapiro mainly concentrated on the investigation of this process and its various applications.<sup>2)</sup> Work was done in two directions: 1) study of elastic moderation of neutrons from a pulsed source in a heavy (mean mass number of the nuclei  $A \gg 1$ ) medium; 2) study of the thermalization and diffusion of thermal neutrons in various media and systems. The study of the first direction made it possible to develop and realize an original method of spectrometry of neutron-induced reactions: spectrometry based on measurement of the neutron moderation time in lead (see also the paper of Yu. P. Popov in this journal issue, which will be cited in what follows as I). The second direction of study made it possible to find some important general properties of the interaction of slow neutrons with matter and opened up considerable possibilities for developing new methods of measuring the neutronphysics parameters of matter, nondestructive element analysis of samples and media (in particular, in nuclear geophysics for analysis of core samples).

This paper is devoted to an exposition of these studies of Shapiro, their further development, and the present status of such directions in neutron physics. The paper consists of four sections. Section 1 is introductory in nature. In it, we formulate the basic concepts and define the quantities that characterize neutron transport in matter, discussing the laws that govern this process and its main stages; we derive the kinetic equation in the theory of neutron transport and the most important methods of its approximate solution. In Sec. 2, we describe Shapiro's studies of the parameters of nuclear chain reactions in uranium-graphite multiplying systems. Sections 3 and 4 are devoted to investigation of nonstationary moderation and diffusion of neutrons, respectively.

#### 1. BASIC CONCEPTS AND DEFINITIONS. THE KINETIC **EQUATION IN THE THEORY OF NEUTRON** TRANSPORT

#### 1. Neutron distribution functions

The neutrons produced as a result of nuclear reactions usually have energies of the order of tens of kilo-electronvolts and higher. Colliding with the nuclei of matter, such neutrons gradually lose their energy (are moderated) and move through space (they diffuse). To describe a field of neutrons in the process of their moderation and diffusion. and also the effects due to their interaction with matter, it is convenient to introduce the concept of the neutron distribution function  $f(\mathbf{r}, E, \mathbf{\Omega}, t)$  at the time t in the "phase space" characterized by the spatial coordinates  $\mathbf{r}$ , the energy E, and the unit vector  $\Omega$  in the direction of motion of a neutron; the distribution function has the following physical meaning. The quantity  $fdEd\Omega$  is the number of neutrons in the interval of energies (E,E+dE) in the element of solid angle  $d\Omega$ that in unit interval of time at the time t and the point  $\mathbf{r}$ intersect a unit area normal to the vector  $\Omega$ . It follows from this that the function n = f/v ( $v = \sqrt{2E/m_n}$  is the neutron velocity) is the phase density of the neutrons, i.e.,  $n(\mathbf{r}, E, \Omega, t) d\mathbf{r} dE d\Omega$  is the number of neutrons at the time t in the element of "phase space"  $d\mathbf{r}dEd\Omega$ . Any physical quantity that is proportional to the intensity of the neutron field can be represented as a linear functional of f. Moreover, as a rule, these functionals reduce to functionals of "simpler" functions:<sup>3)</sup>

$$\Phi_0(\mathbf{r},E,t) = \frac{1}{4\pi} \int d\mathbf{\Omega} f(\mathbf{r},E,\mathbf{\Omega},t)$$

and

$$\mathbf{\Phi}_{1}(\mathbf{r},E,t) = \frac{1}{4\pi} \int d\mathbf{\Omega} f(\mathbf{r},E,\mathbf{\Omega},t)\mathbf{\Omega}; \tag{1}$$

the scalar  $\Phi_0$  is the space-energy distribution of the neutron flux density (in what follows, we shall refer to this for brevity as the neutron flux density), while the vector  $\Phi_1$  is the space-energy distribution of the neutron current density. For example, the number of neutrons with energy in the interval (E,E+dE) that in unit time cross at the point  $\mathbf{r}$  the unit area normal to the unit vector  $\mathbf{v}$  in the positive direction of this vector is

$$dN_{\nu}(\mathbf{r},E,t) = dE \left[ \frac{1}{4} \Phi_0(\mathbf{r},E,t) + \frac{1}{2} \nu \Phi_1(r,E,t) \right];$$
 (2)

the mean path traversed by the neutrons per unit time within some volume V is

$$L_V = \int_V d\mathbf{r} \int dE \Phi_0(\mathbf{r}, E); \tag{3}$$

the number of reaction events induced per unit time by neutrons in the nuclei of some isotope X is

$$N_{\mathbf{r},X} = \int d\mathbf{r} n_X(\mathbf{r}) \int dE \Phi_0(\mathbf{r}, E) \sigma_X(E)$$
$$= \int_V d\mathbf{r} dE \Phi_0(\mathbf{r}, E) \Sigma_X(\mathbf{r}, E), \tag{4}$$

where  $n_X(\mathbf{r})$  is the density of the nuclei of isotope X at the point  $\mathbf{r}$ , and  $\sigma_X(E)$  is the cross section of the considered reaction; the integral over  $d\mathbf{r}$  is over the complete volume V occupied by the sample containing the isotope X,

$$\sum_{\mathbf{Y}}(\mathbf{r},E) \equiv n_{\mathbf{Y}}(\mathbf{r})\sigma_{\mathbf{Y}}(E) \tag{5}$$

is called the macroscopic cross section of this reaction, and its reciprocal,  $l_X(\mathbf{r},E)=1/\Sigma_X(\mathbf{r},E)$ , is called the corresponding mean range of a neutron with energy E.

#### 2. Kinetic equation. Collision integral

The distribution function f satisfies the linear Boltzmann kinetic equation (see, for example, Ref. 5)

$$\frac{1}{n}\frac{\partial f}{\partial t} = -\mathbf{\Omega}\nabla f + \hat{I}[f] + Q. \tag{6}$$

Here  $\hat{I}[f]$  is the so-called collision integral, which determines the change of the function f (more precisely, f/v) as a result of collisions with atoms of the medium, and Q is the density of the neutron sources [i.e.,  $Q(\mathbf{r},E,\mathbf{\Omega},t)d\mathbf{r}dEd\mathbf{\Omega}$  is the number of neutrons formed per unit time at the time t in the element of phase space  $d\mathbf{r}dEd\mathbf{\Omega}$ ]. Equation (6) means that the variation in time of the phase density of the neutrons at any point  $\mathbf{r}$  at time t (left-hand side of the equation) is equal to its change as a result of the displacement of the neutrons in space, described by  $-\mathbf{v}\nabla(f/v)$ , as a result of the collisions of the neutrons, the collision integral, and the supply of neutrons from the sources.

The collision integral can be represented in the form

$$\hat{I}[f] = \int dE' d\mathbf{\Omega}' f(\mathbf{r}, E', \mathbf{\Omega}', t) \mathbf{\Sigma}_{incl}(\mathbf{r}, E', \mathbf{\Omega}' \to E, \mathbf{\Omega})$$

$$-f(\mathbf{r},E,\mathbf{\Omega},t)\Sigma_{t}(\mathbf{r},E). \tag{7}$$

Here  $\Sigma_{\text{incl}}(\mathbf{r}; E', \mathbf{\Omega}' \rightarrow E, \mathbf{\Omega})$  is the macroscopic [i.e., in accordance with (5), multiplied by the density  $n(\mathbf{r})$  of nuclei in the medium] inclusive cross section in the laboratory system for the scattering into unit interval of energies near E and unit solid angle near  $\Omega$  of a neutron that before the interaction (at the point r) had energy E' and direction of motion  $\Omega'$ , and  $\Sigma_{r}(\mathbf{r},E)$  is the total macroscopic cross section for interaction at the point  $\mathbf{r}$  of a neutron with energy E. In the range of neutron energies in which we are interested (≤10 MeV), the cross section  $\Sigma_{\text{incl}}(\mathbf{r}; E', \Omega' \rightarrow E, \Omega)$  in the general case is a sum of the macroscopic cross sections of elastic,  $\Sigma_{\rm el}({\bf r}; E', {\bf \Omega}' \rightarrow {\bf \Omega}, E)$ , and inelastic,  $\Sigma_{\rm in}({\bf r}, E', {\bf \Omega}' \rightarrow E, {\bf \Omega})$ , scattering and fission  $\Sigma_{t}(\mathbf{r}; E', \Omega' \rightarrow E, \Omega)$ ; if the medium contains deuterium or beryllium, then at energy  $E \gtrsim 2$  MeV a small contribution to  $\Sigma_{incl}$  may also be made by reactions of the type (n,2n) (multiplication on fast neutrons). The total cross section  $\Sigma_{\ell}(\mathbf{r},E)$  contains, besides the total (integrated over the final states) cross sections of the listed processes, the macroscopic cross section  $\Sigma_c(\mathbf{r},E)$  of neutron absorption (capture), absorption being understood here as both radiative capture and other reactions in which neutrons are not produced.

The quantities that are the reciprocals of the macroscopic cross sections are the mean ranges of the neutrons until the corresponding interaction. For example,  $l_c(\mathbf{r},E) \equiv 1/\Sigma_c(\mathbf{r},E)$  is the range of a neutron until capture, and  $l_t(\mathbf{r},E) \equiv 1/\Sigma_t(\mathbf{r},E)$  is the total range of a neutron with energy E near the point  $\mathbf{r}$ .

# 3. Stages in the "life of an average neutron in the medium"

We consider qualitatively the "history of the life of an average neutron in the medium," i.e., the evolution in time of the solution of Eq. (6) in the case of a pulsed source of fast neutrons. For simplicity, we shall assume at first that we have a nonmultiplying medium (it does not contain fissioning isotopes).

At high energies (more precisely, at  $E > E_{\rm th}$ , where  $E_{\rm th}$  is the threshold of inelastic scattering of neutrons by the nuclei of the medium), the main contribution to the collision integral is made by elastic and inelastic scattering; absorption plays a small role. This is the region of inelastic moderation of the neutrons. After several collisions, the neutrons "slip out" of this region without, as a rule, having moved far from the place where they were produced. Therefore, f is appreciably nonvanishing only at distances from the source of order  $l_t$ . However, already after the first collision (except in the case of neutron scattering by a proton) f depends weakly on  $\Omega$ . Further, the neutron energy spectrum rapidly becomes softer with the time t that has elapsed from the neutron pulse.

For  $E < E_{th}$ , the moderation occurs only as a result of elastic interaction of the neutrons with the nuclei of the medium: elastic scattering by free nuclei at rest when the energies are large compared with the energy of chemical binding and the thermal motion of the atoms  $(E \ge 0.1-1.0 \text{ eV})$  and

thermalization (approach to thermal equilibrium with the medium) at lower energies. In this region, the mean energy loss in one collision becomes much smaller, especially in the case of a heavy medium. A neutron can diffuse in the medium for a long time before it becomes thermal. During this time, a significant fraction of the neutrons can be absorbed despite the relatively small cross section for radiative capture. This region of energies is usually called the region of neutron radiative capture. Here the energy spectrum of the neutrons as a function of t is a bell-shaped curve (with a relatively small width in the case of a heavy medium; see Sec. 3), the maximum of which is shifted to the region of small energies at a rate  $\sim t^{-2}$ . As the energy decreases, the spatial region in which f is appreciably nonzero expands, and the dependence of f on  $\Omega$  becomes even weaker.

After the stage of moderation has been completed (at the end of thermalization), the process of diffusion of the thermal neutrons in the medium commences. The neutron spectrum becomes nearly Maxwellian with a temperature equal to the temperature of the medium, and Eq. (6) takes a form analogous to the equation of spatial diffusion (subsection 6) with sources in the form of the distribution of the moderated neutrons. The thermal neutrons diffuse in the medium until they are captured or escape from it.

In the case of a multiplying medium, this scenario is augmented by a continuous additional supply of fast fission neutrons. Near the position of fissioning isotopes, there will, even in the case of a pulsed source, be neutrons of all energies simultaneously, from thermal to the fission energy, and even in the case of a pulsed source the neutron spectrum will depend weakly on the time.

# 4. Initial and boundary conditions. The existence and uniqueness of a solution of the kinetic equation

Equation (6) always has one and only one solution when  $t>t_0$  if the boundary and initial conditions are correctly formulated. If the medium is unbounded, the correct boundary condition is, for example, the requirement that  $r^2f$  be bounded as  $r\to\infty$ . In the case of a bounded nonconcave medium, it is necessary to specify on its surface f, for  $\Omega \cdot \nu \leq 0V$ , where  $\nu$  is the outer normal to the surface. Specification of the initial condition means specification of f everywhere within the medium at the time  $t_0$ .

Suppose that the neutron sources, and therefore f, do not depend on the time and the medium is nonmultiplying. Then a solution of Eq. (6) exists and is unique if the boundary conditions are properly posed. The existence and uniqueness of a solution of (6) in the case of a steady source in a multiplying medium depend on the values of the effective coefficient of neutron multiplication  $k_{\rm eff}$  (see Sec. 2). If  $k_{\rm eff} > 1$ , then as a result of action of the source as  $t \to -\infty$  infinitely many neutrons are produced in the medium, and Eq. (6) becomes meaningless.

#### 5. Diffusion approximation

A universal method of calculating the function f is the Monte Carlo method. However, in a number of practically important cases use of this method, even with modern fast

computers, requires extremely long computing times. Therefore, approximate methods of solving Eq. (6) play an important role (especially for elucidating the various features of the physics of neutron transport). For example, the diffusion approximation is very widely used. It is based on the fact that in many cases the function f depends weakly on  $\Omega$  and can be represented approximately in the form

$$f(\mathbf{r}, E, \mathbf{\Omega}, t) = \frac{1}{4\pi} \left[ \Phi_0(\mathbf{r}, E, t) + 3\mathbf{\Omega}\Phi_1(\mathbf{r}, E, t) \right]$$
(8)

[see Eq. (1)] with  $\Phi_0 \gg |\Phi_1|$ . When this expression is substituted in (6), one ignores (or takes into account only approximately) the quantities of order  $|\Phi_1|$  compared with  $\Phi_0$ ; in particular, this applies to the term  $\partial \Phi_1/\partial t$ . Then in the case of an isotropic source (Q does not depend on  $\Omega$ ), it is possible to obtain the following system of equations, which in conjunction with (8) determines the function f:

$$\left(\frac{1}{v}\frac{\partial}{\partial t} - \frac{l_{tr}}{3}\Delta + \Sigma_{t}\right)\Phi_{0} = \int dE' \Phi_{0}(\mathbf{r}, E', t)\Sigma_{incl}(E')$$

$$\rightarrow E) + 4\pi Q, \tag{9}$$

$$\boldsymbol{\Phi}_1 = -\frac{1}{3} l_{tr} \boldsymbol{\nabla} \boldsymbol{\Phi}_0 \tag{10}$$

[note that (10) is Fick's law]. Here  $l_{\rm tr}$  is the neutron transport range:

$$l_{\rm tr} = 1/[\Sigma_t - (\Sigma_{\rm el} + \Sigma_{\rm in}) \overline{\cos \theta}], \tag{11}$$

and  $\cos \theta$  is the mean cosine of the angle of scattering (the sum of elastic and inelastic), and

$$\Sigma_{\text{incl}}(E' \to E) = \int d\mathbf{\Omega} \Sigma_{\text{incl}}(E', \mathbf{\Omega}' \to E, \mathbf{\Omega}). \tag{12}$$

Equation (9) is valid only in the case of a homogeneous medium. If there are several regions that are each occupied by a homogeneous medium, then on their interfaces  $\Phi_0$  and  $\Phi_1 \cdot \nu$ , where  $\nu$  is the normal to the interface, must be continuous. On a nonconcave boundary with vacuum, under the assumption that all the sources are situated within the medium, it is necessary to require vanishing of the number of neutrons incident from without on this boundary, i.e., in accordance with (2), it is necessary to set

$$\Phi_0(\mathbf{r},E) - \frac{2}{3}l_{tr}(E)\boldsymbol{\nu}\nabla\Phi_0(\mathbf{r},E)\big|_{\mathbf{r}=\mathbf{r}_s} = 0$$
 (13)

 $(\mathbf{r}_s)$  is any point on the boundary of the medium). Alternatively, assuming that near the boundary  $\Phi_0$  decreases linearly along the normal to it, this condition can be replaced by the approximate condition  $\Phi_0(\mathbf{r}_{\text{extr}}, E) = 0$ , where  $\mathbf{r}_{\text{extr}}$  is any point on an extrapolated boundary, which is a surface separated from the boundary of the medium in its exterior by the extrapolation length  $z_{\text{extr}} = (2/3)l_{\text{tr}}$  (more precisely,  $z_{\text{extr}} = 0.71l_{\text{tr}}$ ). This greatly facilitates the solution of Eq. (9). For example, in the case of a homogeneous medium filling a bounded volume, this makes it possible to seek  $\Phi_0$  as an expansion with respect to an orthonormal system of eigenfunctions of the Laplacian (harmonics)  $R_n(\mathbf{r})$  that vanish on the extrapolated boundary  $[R_n(\mathbf{r}_{\text{extr}}) = 0]$ :

$$\Phi_0(\mathbf{r}, E, t) = \sum_{n=0}^{\infty} \varphi_n(E, t) R_n(\mathbf{r}_n), \tag{14}$$

$$\Delta R_n(\mathbf{r}) + B_n^2 R_n(\mathbf{r}) = 0, \quad \int d\mathbf{r} R_n(\mathbf{r}) R_m(\mathbf{r}) = \delta_{nm}.$$
 (15)

The lowest eigenvalue  $B_0^2$  corresponding to the ground harmonic  $R_0(\mathbf{r})$  is called the geometric parameter. Substituting (14) in (9), we obtain for  $\varphi_n$  the equations

$$\left(\frac{1}{\upsilon}\frac{\partial}{\partial t} + \frac{l_{\text{tr}}}{3}B_n^2 + \Sigma_t\right)\varphi_n$$

$$= \int dE' \varphi_n(E',t)\Sigma_{\text{incl}}(E' \to E)$$

$$+ 4\pi \int d\mathbf{r}R_n(\mathbf{r})Q(\mathbf{r},E,t). \tag{16}$$

### 6. "Single-velocity" approximation

A radical simplification in the description of the diffusion of thermal neutrons is made possible by the assumption that these neutrons are in thermal equilibrium with the medium, i.e.,

$$\Phi_0(\mathbf{r}, E, t) = n(\mathbf{r}, t)vM_T(E), \tag{17}$$

$$M_T(E) = \frac{1}{\sqrt{\pi}} \frac{1}{T^{3/2}} \sqrt{E} e^{-E/T},$$
 (18)

where  $n(\mathbf{r},t)$  is the density of the neutrons at the point  $\mathbf{r}$  at the time t, and T is the temperature of the medium. If at the same time the medium is nonmultiplying (i.e.,  $\Sigma_{\text{incl}} = \Sigma_S$ , the scattering cross section), then by the principle of detailed balance

$$vM_{\tau}(E)\sum_{s}(E \rightarrow E') = v'M_{\tau}(E')\sum_{s}(E' \rightarrow E)$$
 (19)

and Eq. (9) is readily reduced to the form<sup>4)</sup>

$$\frac{\partial n}{\partial t}$$
  $-D\Delta n + \frac{1}{T_0}n = 4\pi\langle Q \rangle_T$ ,

$$D = \frac{1}{3} \langle v l_{\text{tr}} \rangle_T, \quad \frac{1}{T_0} = \langle v / l_c \rangle_T, \tag{20}$$

where the symbol  $\langle ... \rangle_T$  denotes averaging over  $M_T(E)$ . Here D and  $T_0$  are called the diffusion coefficient and the lifetime, and  $L = \sqrt{DT_0}$  is the diffusion length of the thermal neutrons.

Let the medium by unbounded and a point "unit" source be situated at the origin. Then if the source is stationary,  $4\pi\langle Q\rangle_T = \delta(t)\,\delta(\mathbf{r})$ , the solution of Eq. (19) has the form

$$n = \frac{1}{4\pi Dr} e^{-r/L}. (21)$$

Then for the mean square of the displacement of a neutron before absorption, we have

$$\overline{r^2} = \int d\mathbf{r} r^2 n(\mathbf{r}) / \int d\mathbf{r} n(\mathbf{r})$$

$$= \int_0^\infty dr \ r^3 \ e^{-r/L} / \int_0^\infty dr \ r \ e^{-r/L} = 6L^2, \tag{22}$$

i.e.,  $L^2$  has the meaning of  $(1/6)\overline{r^2}$ . In the case of a pulsed source,  $4\pi\langle Q\rangle_T = \delta(t)\,\delta(\mathbf{r})$ , and in the absence of absorption  $(I_c = \infty)$ , we have

$$n = \frac{1}{(4\pi Dt)^{3/2}} \exp\left(-\frac{r^2}{4Dt}\right),\tag{23}$$

$$\overline{r^2(t)} = \int d\mathbf{r} r^2 n(\mathbf{r}, t) / \int d\mathbf{r} n(\mathbf{r}, t) = 6Dt.$$
 (24)

In the case of a bounded homogeneous medium, the coefficients  $\varphi_n$  of the expansion of the neutron flux density with respect to the functions  $R_n$ , i.e., the solutions of Eqs. (19), can also be readily found in the stationary and pulsed cases.

### 7. Age approximation

Another case of frequent use of the diffusion approximation is the description of neutron transport in a heavy non-multiplying medium in the process of elastic moderation under the assumption that the nuclei are free and at rest, i.e.,  $\Sigma_{\text{incl}} = \Sigma_{\text{el}}$ . Then the macroscopic cross section for scattering of a neutron by nuclei with mass number  $A \gg 1$  is nonzero only in a narrow range of final energies,  $\alpha^2 E' < E < E'$ ,  $\alpha = (A-1)/(A+1)$ , and in this range it is equal to

$$\Sigma_{el}(E' \to E) = (A+1)^2 \Sigma_{el}(E')/4AE'.$$
 (25)

Therefore, the integral on the right-hand side of (9) can be calculated approximately. For this, we go over from the energy to a new variable, the "age"  $\tau$  of the neutrons, and from the function  $\Phi_0$  to the moderation density  $q(\mathbf{r},\tau,t)$  (i.e., the number of neutrons per unit volume near  $\mathbf{r}$  that go over at the time t in unit time from the range of ages less than  $\tau$  to the range of ages greater than  $\tau$ ) in accordance with the expressions

$$\tau = \frac{1}{3\zeta} \int_0^u du' l_s(u') l_{tr}(u'), \quad q = \frac{1}{\zeta l_s(u')} \Phi_0, \quad (26)$$

where  $u = \ln(E_M/E)$  is the so-called lethargy  $(E_M$  is the maximum energy of the neutrons from the source), and

$$\zeta = 1 - \frac{(A-1)^2}{2A} \ln \frac{A+1}{A-1} \tag{27}$$

is the mean logarithmic loss of energy in a collision. Then expanding in the integrand q(u') in a series in powers of u'-u and restricting ourselves to the first two terms of the expansion, we readily obtain the equation

$$\frac{3}{v l_{tr}} \frac{\partial q}{\partial t} + \frac{\partial q}{\partial \tau} - \Delta q + \frac{3}{l_{tr} l_{c}} q = \frac{3\zeta}{l_{tr} l_{c}} Q. \tag{28}$$

In the case of a stationary source  $(\partial q/\partial t=0)$ , Eq. (28) has the same form as Eq. (19), and (apart from the constant factor  $D^{-1}$ )  $\tau$  plays the role of the time. This is why it became known as the "age of the neutrons." By virtue of this analogy, this equation can be solved by means of the same methods as Eq. (19). We shall consider the solution of Eq. (28) with a nonstationary source in Sec. 3.

#### 2. STUDY OF MULTIPLYING MEDIA

# 1. Nuclear fission chain reactions. The neutron multiplication coefficient

We recall the basic properties of a multiplying medium, i.e., a medium that contains fissioning isotopes, so that in it there can be a nuclear fission chain reaction. (For more details, see Ref. 5). We restrict ourselves to the case of the fission of uranium [natural:  $^{235}U(0.72\%)+^{238}U$ ]; this case was investigated by Shapiro. In uranium, fission occurs almost entirely as a result of the capture of a thermal neutron by a  $^{235}U$  nucleus, and the mean number of neutrons (with energy  $\approx 1$  MeV) for each thermal neutron captured in uranium is

$$\eta = \rho \nu = 1.33, \tag{29}$$

where  $p\approx0.545$  is the probability of fission as a result of such capture, and  $\nu=2.44$  is the mean number of neutrons resulting from fission of the <sup>235</sup>U nucleus by a thermal neutron. In an unbounded homogeneous medium, the coefficient of neutron multiplication [the number of thermal neutrons of the (n+1)th generation for thermal neutron of the nth generation] is determined by the "four-factor formula"

$$k = \eta \varepsilon \varphi \theta \tag{30}$$

(in Shapiro's papers, the product  $\eta \varepsilon$  was denoted by the symbol  $\nu$ , as was then standard). Here  $\varepsilon$  is the coefficient of multiplication on fast neutrons ( $\varepsilon - 1 \sim 10^{-2}$ ),  $\varphi$  is the probability of avoiding radiative capture in the process of moderation of a neutron to the thermal energy, and  $\theta$  is the probability of capture of a thermal neutron by uranium. In the case of a bounded medium (system), the effective multiplication coefficient is  $k_{\text{eff}} = k(1-P)$ , where P is the probability of escape of the neutrons from the medium. Systems with  $k_{\text{eff}} > 1$ ,  $k_{\text{eff}} = 1$ , and  $k_{\text{eff}} < 1$  are called supercritical, critical, and subcritical, respectively. Pure uranium is a subcritical system because of the small value of  $\varphi$ . To ensure the fastest passage through the "dangerous" stage of elastic moderation, a moderator (for example, water or graphite) is added to the system. In what follows, to be specific, we shall consider uranium-graphite systems, which were investigated by Shapiro. For any homogeneous uranium-graphite system, k < 1 also. To raise the value of k (by an increase of  $\varphi$ ), heterogeneous systems are used. Since in this case too k-1is small (<0.1), an important problem (up to now!) has been the reliable determination of the factors in the expression (30) and the study of their dependence on the parameters of the system. One of the most effective ways of solving this problem is to study a field of neutrons in subcritical systems with a local stationary source.

### 2. Subcritical systems. The prism method

By 1947 (by the beginning of Shapiro's studies), it was already known (see, for example, Ref. 6) that in systems of uranium-graphite type the transport of thermal neutrons can be described in the single-velocity approximation [see (20)] with neglect of the change in n during the time of moderation:

$$\frac{\partial n}{\partial t} - D\Delta n + \frac{1}{T_0} n = Q_0(\bar{r}, t) + \frac{k}{T_0} \int d\bar{r}' n(\bar{r}') w(\bar{r}' - \bar{r}), \tag{31}$$

where  $Q_0(\bar{r},t)$  is the external source, and  $w(\bar{r}'-\bar{r})$  is the distribution (normalized to unity) with respect to  $\bar{r}$  of a fission neutron produced at the point  $\bar{r}'$  and moderated to thermal energy. In the age approximation [Eq. (28)],

$$w(\bar{r}' - \bar{r}) = (4\pi\tau_T)^{-3/2} \exp[-(\bar{r} - \bar{r}')^2/3\tau_T], \tag{32}$$

where  $\tau_T$  is the age of the thermal neutrons (for graphite with  $\rho$ =1.65 g/cm<sup>3</sup>,  $\tau_T$ =350 cm<sup>2</sup>). These expressions are valid both for a homogeneous system and for a system in the form of a regular lattice if the densities in neighboring cells are nearly the same. For a uranium-graphite system, this condition is practically always satisfied if the size of the uranium blocks is much smaller than the lattice period.

In the case of a pulsed source in a subcritical system, the coefficients of the expansion with respect to the eigenfunctions of the Laplacian of the type (14) are exponentially damped with time, the ground harmonic being damped most slowly. Its decay rate is

$$\lambda_0 = \frac{1 - k_{\text{eff}}}{T_0} = \frac{1}{T_0} \left[ 1 - k + \kappa^2 (L_M^2 \delta + k \tau_T) \right], \tag{33}$$

where  $\kappa^2$  is the geometric parameter (the eigenvalue of the ground harmonic),  $L_M$  is the diffusion length of the thermal neutrons in the pure moderator, and  $\delta = 1 - \theta$  is the fraction of neutrons captured in the moderator. By measuring  $\lambda_0$  for different values of  $\kappa^2$ , it is possible to determine k and the conditions for the system to become critical ( $\lambda_0 = 0$ ):

$$\kappa^2 = \kappa_{\rm cr}^2 \equiv (k-1)/A, \quad A = L_M^2 \delta + k \tau_T, \tag{34}$$

and from this one can determine the size of the critical system ( $\kappa_{cr}^2$  is called the material parameter, and A is the neutron migration area).

In the case of a stationary source, a system of practical interest is one in the form of a rectangular prism for which one of the dimensions—along the z axis—is much greater than the other two. Let its dimensions along the x and y axes be the same and equal to 2a, the neutron source be a point, and this source be placed on the z axis at the point z=0. Then the solution of Eq. (31) that vanishes on the lateral extrapolated boundary of the prism, i.e., on  $x=\pm a_1$ ,  $y=\pm a_1$ , and  $a_1=a+z_{\rm extr}$ , can be represented in the form

$$n = \sum_{l,m=0}^{\infty} C_{l,m}(z) \cos \frac{(2l+1)\pi x}{2a_1} \cos \frac{(2m+1)\pi y}{2a_1}.$$
 (35)

Then far from the source (more precisely, where  $|z| \gg \sqrt{2\tau_T}$ ), the values of  $C_{l,m}(z)$  decrease with |z| as  $\sim \exp(-\alpha_{l,m}|z|)$ , where

$$\alpha_{l,m}^2 = \frac{(2l+1)^2 + (2m+1)^2}{4a_1^2} \pi^2 - \frac{k-1}{L_M^2 \delta + k \tau_T}$$

$$= \frac{(2l+1)^2 + (2m+1)^2}{4a_1^2} \pi^2 - \kappa_{cr}^2. \tag{36}$$

In particular, for  $|z| \ge a_1/2$ , the density of thermal neutrons is  $n \sim \exp(-\alpha_{00}|z|)$ , and by measuring the ratio of the densi-

ties at two different z it is possible to determine  $\alpha_{00}$  and, therefore, the parameter  $\kappa_{cr}^2$ . This method of investigating multiplying systems became known as the exponential method, or the prism method.

In the forties, there were not yet any convenient pulsed neutron sources, and the prism method was one of the two most effective approaches to the study of multiplying systems (see, for example Ref. 8, Chap. 13). An alternative to it was the method of approach to the critical state by increasing the volume of the multiplying medium with simultaneous measurement of the coefficient of multiplication of neutrons from an external source. In some cases, such an approach made it possible to obtain more accurate results but required a much greater amount of the multiplying medium, and in the forties that was a very serious problem.

### 3. Shapiro's studies of multiplying systems

By the time when Shapiro began his studies, Soviet physicists<sup>5)</sup> had already proposed an approach to the calculation of the neutron multiplication coefficient and had made a preliminary analysis (mainly theoretical estimates) of the factors in the expression (30), had established the advantages of a heterogeneous system as a system with a significantly larger value of  $\varphi$ , had considered the effects that could influence the coefficient  $\theta$ , and had developed a general theory of a heterogeneous reactor and methods of measuring neutronphysics parameters, in particular, the prism method. However, because of the absence of reliable experimental data on the neutron cross sections, in particular, on radiative capture in uranium, and the rough nature of the methods of calculation, the theoretical estimates of the parameters of multiplying systems were rather uncertain, while the experimental data were fragmentary (for example, there had been reliable determinations of the diffusion length  $L_C$  for pure graphite and, for some systems,  $\kappa_{cr}^2$ ).

Shapiro's studies (see footnote 2) were devoted to the determination of k and the factors that occur in it, and also the determination of the migration area A for uranium—graphite systems with different uranium concentrations and the influence of the properties of the neutron distribution (in particular, the degree of approximation of the neutrons to thermal equilibrium with the medium) on these quantities.

The main experiments were performed on a large prism  $(180\times180\times420 \text{ cm}^3)$  of graphite bricks measuring 20  $\times 20 \times 60 \text{ cm}^3$  with circular channels ( $\emptyset = 4.4 \text{ cm}$ ) assembled with a displacement in such a way that there were additional square channels  $(6.67 \times 6.67 \text{ cm}^2)$ . Uranium rods  $(\emptyset = 3.5)$ cm) with different concentrations were placed in the channels (one of the forms of arranging the uranium is shown in Fig. 1). The empty channels were covered with graphite plugs. The neutron density was measured (by a boron camera) on the vertical axis of the prism at different distances from an Ra+Be source. For maximum accuracy in the determination of the value of  $\kappa_{\rm cr}^2$ , Shapiro greatly refined the theory of the prism method. He developed a method of accurate calculation of corrections that take into account the different extrapolation lengths for fast and thermal neutrons and the presence at the ends and in the lateral layers of the prism of a material different from the material of the main

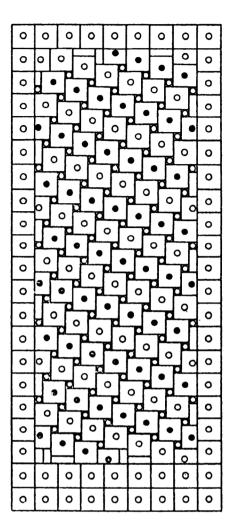


FIG. 1. Layout of a facility (prism) used to study the parameters of a uranium-graphite system, showing one of the arrangements of the uranium blocks (dark circles).

part of the prism. As a result, he achieved an accuracy in the measurement of  $\kappa_{cr}^2$  equal to  $\pm 0.7 \cdot 10^{-5}$  cm<sup>-2</sup>. He proposed and implemented a method of measuring the fraction  $\delta$  of neutrons captured by the graphite by comparing the densities of the thermal and superthermal ("supercadmium") neutrons in a cell; this method later became known as the cadmiumratio method. Shapiro also developed an independent method for measuring  $\delta$  by "poisoning" the prism with a small Cd admixture. Taken together, these modifications made it possible to measure with good accuracy ( $\sim 0.5\%$ ) the absolute value of the coefficient  $\theta$  and (moreover, much more accurately) the influence on it of the various factors. 6) We must mention especially the study of the temperature dependence of the parameters of uranium-graphite systems. This was done by creating a special experimental setup, a prism in a thermal bath in which graphite was heated by an electric current, this being done both uniformly over the cell and by heating individual parts of bricks. Besides the solution of purely technical problems, this required refinement of the theory (in particular, allowance for "crossflow of neutrons through slits formed during the insulation of the bricks). In these experiments, the mean velocities of the thermal neutrons in pure graphite and in uranium-graphite systems were

also measured. It was found that in the graphite the neutrons arrive in complete thermal equilibrium with the medium, while in a uranium-graphite system (with "normal" concentration), the temperature of the neutrons was  $\sim 30\%$  above the temperature of the medium. Using data on  $\delta$  and  $L_C$ , Shapiro determined the migration area A, and then, using the expression (34), k as well. These measurements were made with two concentrations of the uranium rods: normal and half the concentration, for which the value of  $1-\varphi$  (the probability of resonant capture in uranium) also differs by a factor of two. This made it possible to determine separately the factors  $\eta \varepsilon$  and  $\varphi$  in the expression (30) (see also footnote 5). His results showed that the theoretical estimates of  $\varphi$  adopted at that time required serious correction. In this connection, Shapiro developed a theory of resonant capture of neutrons in fat uranium rods (the effective dimensions of which were much greater than the mean neutron range at the resonance energy) that took into account the Doppler broadening of the resonance line and neutron rescattering within a rod. Calculations made earlier that did not take into account these effects gave for the probability of radiative capture of neutrons in cylindrical uranium rods a value ~40% below the experimental value! Allowance for these effects enabled Shapiro to reconcile the theory with the experimental data.

Thus, in this series of studies Shapiro developed methods for determining with high accuracy and reliability the basic parameters of multiplying systems, and by means of them uranium-graphite heterogeneous assemblies, which were extremely important at that time, were studied. We emphasize that even today macroscopic experiments of prism type make it possible to obtain reliable results and test microscopic calculations; the approaches developed by Shapiro are still used, and the experimental data he obtained (for example, for  $L_C^2$  and  $\tau_T$ ) hardly differ from the data recommended today.

# 3. NONSTATIONARY NEUTRON MODERATION IN HEAVY NUCLEI

### 1. Some history

In 1944, in I. V. Kurchatov's seminar, E. L. Feĭnberg showed in the age approximation that the neutrons from a pulsed source in a heavy medium become grouped in velocity in the process of elastic moderation near some mean value, which is different for each instant of time after the neutron pulse. This fact, as was noted by Feĭnberg and L. Lazareva (private communication, 1950), can be used for neutron spectrometry. Shapiro considered this question in detail in a seminar at the P. N. Lebedev Physics Institute in 1950. He showed that the pulsed neutron sources developed by that time on the basis of the D+D and D+T reactions make it possible to implement such a method of neutron spectrometry.

Since, as has already been mentioned, the separate paper I of Yu. Popov in this collection is devoted to neutron spectrometry based on the moderation time (one of the most important directions of Shapiro's scientific work), we restrict ourselves here to the investigations into the physics of the process of moderation of neutrons from a pulsed source in a

heavy medium. At the suggestion of Shapiro, the theory of this process was developed by the present author. The experiments were mainly made by A. Isakov, under the supervision of the author. The first results of these investigations were declassified and published in 1955. <sup>9-11</sup> Studies on the same subject were made independently in Sweden. <sup>12</sup> They were subsequently continued, both at the Lebedev Institute <sup>13-16</sup> and abroad. The various aspects of these investigations are most fully described in the monograph of Ref. 17 and in the studies of Refs. 14-16, in which a complete bibliography is also given (see also Ref. 4, pp. 313 and 397).

# 2. Theory of the moderation of neutrons from a pulsed source in a heavy medium

As we have already mentioned, neutrons from a pulsed source that are being moderated in a heavy medium group themselves near a certain mean velocity  $\bar{v}$ , which decreases with the moderation time t. This "focusing" of the neutrons in velocities can be explained on the basis of the following. The probability of scattering of a neutron during 1 s is  $v/l_s$ , i.e., in the case of a constant range before scattering  $\Delta v/v$ (the mean relative loss of velocity during 1 s) is proportional to the velocity. Therefore neutrons with velocities  $v < \bar{v}$  and  $v > \bar{v}$  lose velocity more slowly and more rapidly, respectively, than a neutron with the mean velocity. As a consequence, they congregate in the region of average velocities. The opposite process is due to the scattering-induced spread of the neutron velocities. Since this latter process decreases with increasing A, the focusing effect is stronger, the heavier the nuclei of the medium.

We consider this process quantitatively in the age approximation at first, restricting ourselves for simplicity to the case of an unbounded homogeneous medium and a uniformly distributed neutron source with density  $Q_0$  that at the time t=0 emitted neutrons with velocity  $v_0$ . Then, with allowance for (26), the solution of Eq. (28) is readily obtained in the form

$$\Phi(v,t) = \frac{Q_0 A l_s}{v} \exp\left[-A \int_v^{v_0} dv' \frac{l_s}{v' l_c(v')}\right]$$

$$\times \delta\left(t - A \int_v^{v_0} dv' \frac{l_s}{v'^2}\right)$$
(37)

(we have here used the fact that for  $A \gg 1$  the mean logarithmic energy loss is  $\zeta \approx 2/A$ ), i.e., all the neutrons at each time t have the same velocity  $\bar{v}(t)$ , which in the case of a constant range  $l_s$  is

$$\bar{v}(t) = \frac{Al_s}{t + l_s A/v_0}. (38)$$

If the neutrons from the source had different velocities  $v_{01}$  and  $v_{02}$ , then at the time t the relative difference between their velocities will be

$$\frac{\bar{v}_2(t) - \bar{v}_1(t)}{\bar{v}_2(t) + \bar{v}_1(t)} = \frac{l_s A(1/v_{01} - 1/v_{02})}{2t + l_s A(1/v_{01} + 1/v_{02})} \xrightarrow[t \to \infty]{} 0.$$
 (39)

Thus, in the framework of the age approximation the neutrons "forget" their initial velocity distribution during the

process of elastic moderation, and in the limit  $t \rightarrow \infty$  their spectrum tends to an infinitesimally narrow line. However, for a correct calculation of the dependence of the neutron spectrum on t (i.e., the theoretical resolution of a moderation-time spectrometer), a more rigorous treatment is required.

At the beginning of the studies at the Lebedev Institute, it was known (see the review of Ref. 18 and Ref. 19) that in a nonabsorbing medium with  $l_s$ =const consisting of nuclei of a single species  $\Phi$  in the limit  $t \rightarrow \infty$  tends to a function of just the one variable  $x = vt/l_s$ ; the moments of this function had been found, and an expression whose moments did not differ strongly from the exact ones had been "constructed." However, the degree of error of this expression had not been determined. The spatial dependence of  $\Phi$  had not been considered.

The theory developed in Refs. 11 and 13 showed that in the above case  $\Phi$  can be represented in the form of an expansion

$$\Phi = \operatorname{const} \cdot \exp \left\{ \frac{A+1}{A} f_{-1} \left[ \frac{\bar{v}(t)}{v} \right] + f_0 \left[ \frac{\bar{v}(t)}{v} \right] + \frac{2}{A+1} f_1 \left[ \frac{\bar{v}(t)}{v} \right] + \cdots \right\}, \quad \bar{v}(t) = \frac{A+1}{t} l_s, \quad (40)$$

where the functions  $f_i$  do not depend explicitly on A. For  $f_{-1}$ ,  $f_0$ , and  $f_1$  analytic expressions were found (these being sufficient for the calculation of  $\Phi$  with good accuracy even in the case A=2); because they are cumbersome, we do not give them here. For a sufficiently heavy moderator  $(A \gg 1)$ , a good approximation is the expression

$$\Phi = \operatorname{const} \cdot \exp \left[ -\frac{3A}{4} \left( \frac{\bar{v}}{v} - 1 \right)^{2} \right], \tag{41}$$

which is obtained if the terms proportional to  $f_0$ ,  $f_1$ , etc., are ignored and  $f_{-1}$  is replaced by the first term of its expansion in powers of  $\bar{v}/v-1$ . This means that at each instant the neutron velocity distribution is nearly Gaussian around  $\bar{v}$ v-1 with variance  $\Delta=2/3A$ . If the moderator consists of a mixture of heavy nuclei of different masses, the neutron distribution still remains nearly Gaussian with the mean velocity being determined by the expression (40) with A replaced by  $A = 1/(\sum_{\alpha} \delta_{\alpha}/A_{\alpha})$ , where  $\delta_{\alpha} = l_s/l_{s\alpha}$  is the ratio of the total neutron range to the range until scattering by a nucleus of species  $\alpha$ . At the same time, because of the additional spread velocities in each scattering event variance is increased (the "focusing" is less strong):  $\Delta = (2/3) \sum_{\alpha} \delta_{\alpha} \bar{A} / A_{\alpha}^2$ .

An admixture of hydrogen significantly changes the nature of the neutron spectrum, since when a neutron collides with a proton it can with equal probability acquire a lower energy and leave the region of mean velocities, moving far into the region of low velocities. This produces a "tail" of slow neutrons that decays weakly with decreasing velocity. If the initial neutron spectrum is broad (as is usually the case after the stage of inelastic moderation has been completed), then it will become narrower in accordance with the expressions (38) and (39) until a distribution of the type (41) is established. Allowance for capture and for the dependence of

 $l_s$  on v showed that in cases of practical interest these effects do not have a significant influence on the width of the neutron spectrum. At low energies, the thermal motion and chemical binding of the atoms of the medium begin to affect the neutron spectrum. When  $E \gg K_T$ , where  $K_T$  is the mean kinetic energy of the thermal motion of the atoms, the variance is

$$\Delta = \frac{2}{3A} + \frac{K_T}{6E}.\tag{42}$$

Thus, the relative effect of the thermal motion and the chemical binding on  $\Delta$  has the order of magnitude  $AK_T/E$ ; the effect on the mean velocity is much less, of order  $K_T/E$ .

Because the neutron spectrum is nearly Gaussian, it can be found by calculating the first two reciprocal moments of the neutron velocity distribution directly from an equation of the type (16) and from them determining the parameters of the distribution. Calculations made independently by this method by Shapiro were almost identical to the results of the more rigorous theory.

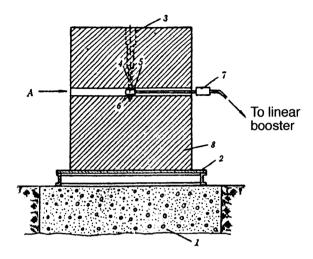
If the source is local, then the neutron distribution in space, in velocity, and in time,  $\Phi_0(\mathbf{r}, v, t)$ , can be represented approximately for  $A \gg 1$  at not too large distances from the source (in the region where the age approximation "works") in the form

$$\Phi_0(\mathbf{r}, v, t) = \Phi(v, t) R[\mathbf{r}, \tau(\bar{v})], \tag{43}$$

where  $\Phi(v,t)$  is the distribution of the neutrons considered above with respect to v and t, and  $R[\mathbf{r},\tau(\bar{v})]$  is the solution of the age equation (28) with stationary source, in which  $\tau$  must be taken to correspond to a velocity equal to  $\bar{v}(t)$ . At large distances from the source, the neutron spectrum at all t is harder and has a much smaller variance. This is due to the fact that the only neutrons that can reach large distances are those that move along nearly straight trajectories, being scattered mainly through small angles, and therefore lose less energy in the collisions.  $^{17,20}$ 

# 3. Experimental investigations of the spectra of neutrons from a pulsed source in a heavy medium

The measurements were made by means of resonance detectors. Into a prism of the investigated moderator of the type shown in Fig. 2, materials possessing isolated resonances with width  $\Gamma$  small compared with the width of the instantaneous neutron spectrum were introduced, and the time dependence  $I_{\nu}(t)$  of the intensity of the captured  $\gamma$  rays was measured. In the first approximation (the refinement is trivial),  $I_{\gamma}(t) \sim \Phi(v_0, t)$ , where  $v_0$  is the velocity corresponding to resonance. Such experiments were performed for lead (the most convenient), graphite, and iron. They showed that the theory correctly predicts the mean time of moderation to a given velocity (Fig. 3) and, in graphite and iron, also the line shape of the neutron spectrum (Figs. 4a and 4b). In lead (Fig. 4c) the experimental FWHM was  $(\delta v/\bar{v})_{\rm exp} = 0.17$ , whereas for pure lead  $(\delta v/\bar{v})_{\rm theor} = 0.135$ . In addition, the experimental neutron distribution had a small "tail" in the low-energy region. This result could be explained under the reasonable assumption that the lead con-



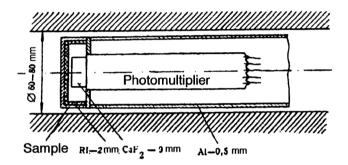


FIG. 2. Arrangement of a lead prism for studying the nonstationary moderation of neutrons: 1) concrete foundation; 2) steel frame; 3) channel for the target tube; 4) target; 5) counhter; 6) sample; 7) preliminary booster; 8) lead prism. The lower figure shows, on a magnified scale, the layout of the scintillation detector and the sample in the channel of the lead prism.

tained as impurities small amounts of hydrogen  $(3.5 \cdot 10^{-4})$  atoms of H per 1 atom of Pb) and oxygen  $(6 \cdot 10^{-3})$  atoms of O per 1 atom of Pb). The measurement of the decrease in time of the density of the integrated neutron flux in the lead prism showed that the theory also satisfactorily described spatial effects (more precisely, the rate of escape of neutrons from the moderator).

These investigations were developed further in connection with problems relating to the improvement of the moderation-time spectrometer (see I).

# 4. NONSTATIONARY THERMALIZATION AND DIFFUSION OF THERMAL NEUTRONS

### 1. Some more history

Simultaneously with the investigations of nonstationary neutron moderation, Frank and Shapiro developed at the P. N. Lebedev Physics Institute studies of neutron transport from pulsed sources in other systems: small uranium—graphite piles (by the method explained in Sec. 2)<sup>21</sup> and pure moderators.<sup>22</sup> The latter proved to be particularly informative. They made it possible to establish some very subtle details of the "life" of neutrons in a medium. Already in the first experiments with blocks of pure moderator of restricted volume performed at the Lebedev Institute and independently by a Swedish group<sup>12</sup> there was found to be a discrep-

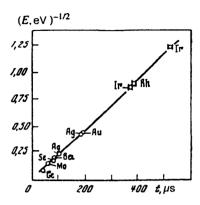


FIG. 3. Relationship between the neutron energy E and the mean moderation time t to reach this energy. The isotopes used for the measurements<sup>14</sup> are shown

ancy with the theory based on the assumption that the neutrons, having come into thermal equilibrium with the medium, have a Maxwellian spectrum with temperature equal to the temperature of the medium (see Sec. 4.2). These experiments could be explained qualitatively (independently by Frank<sup>22,23</sup> and von Dardel<sup>12</sup>), and then in the framework of the systematic phenomenological theory developed by Shapiro, Stepanov, and the present author<sup>24,25</sup> (Sec. 4.3). Thus they discovered the effect of "diffusion cooling" of neutrons-a change in the equilibrium spectrum of neutrons through escape from blocks of matter of finite volume. As a result of these investigations and some studies of foreign authors, 7) the pulsed method received a serious theoretical basis and became the simplest cheap and accurate method of measuring the macroscopic parameters of the interaction of thermal neutrons with matter; it has been widely used in many laboratories throughout the world.

However, this was not the end of the history of investigations into the behavior of thermal neutrons from a pulsed source in finite blocks of a moderator. It was found that in sufficiently small blocks the neutrons do not come into equilibrium with the medium at all (Sec. 4.4). However, subsequently this problem too could be solved. At the same time, new and distinctive features in the evolution of the neutron spectrum in a medium were discovered. Indeed, the method of studying matter by means of a pulsed neutron source has found more and more applications with the passage of the years (Sec. 4.5).

# 2. Discovery of diffusion cooling of a thermal neutron spectrum

We now consider what must be the behavior in time of the total density of neutrons from a pulsed source in a finite block of pure moderator if the neutron spectrum is the Maxwellian spectrum (18) with temperature equal to the temperature of the medium, i.e., the solution of Eq. (20) with  $\langle Q_T \rangle \sim \delta(t)$ . Obviously, the expansion of this solution with respect to the eigenfunctions of the Laplacian (15) that vanish on the extrapolated boundary will have the form

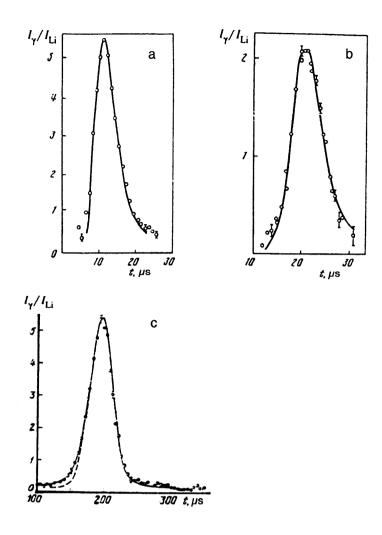


FIG. 4. Graphs of I(t) for a gold sample in graphite (a), iron (b), and lead (c) prisms.<sup>14</sup>

$$n = \sum_{n=0}^{\infty} C_n R_n(\mathbf{r}) e^{-\lambda_n t}, \quad \lambda_n = \frac{1}{T_0} + B_n^2 D, \tag{44}$$

where  $C_n$  are constants determined from the initial distribution of the neutrons. Accordingly, at large t there must be an exponential decrease of n with t with damping constant  $\lambda_0$ . Therefore, by measuring the damping constant of n as  $t \rightarrow \infty$ for different geometric parameters  $B_0^2$  (which in the case of simple forms of the moderator such as a sphere, cylinder, and rectangular parallelepiped can be readily calculated accurately), and plotting the graph of  $\lambda_0$  as a function of  $B_0^2$ , we must obtain a straight line whose intersection with the abscissa determines the neutron lifetime  $T_0$  in the medium, while its slope determines the diffusion coefficient D of the Maxwellian neutrons (in the case of a stationary source, only their product  $DT_0 = L^2$  can be measured). However, as can be seen from the typical graph of this kind for Be in Fig. 5 the dependence of  $\lambda_0$  on  $B_0^2$  deviates strongly from a straight line. Similar results were obtained by the Swedish group who had independently performed such experiments. <sup>12</sup> To explain this effect, Frank <sup>22,23</sup> considered a simple model, <sup>8)</sup> in accordance with which there are in the medium two groups of neutrons with different diffusion coefficients  $D_1$  and  $D_2$ (which therefore escape differently from the medium of finite

volume), and establishment of a Maxwellian spectrum in accordance with the principle of detailed balance occurs over a finite time (as a result of collisions). He then found that the spectra of neutrons in blocks with different  $B_0^2$  will be different, i.e., the escape of the neutrons perturbs their equilibrium spectrum. The dependence of  $\lambda_0$  on  $B_0^2$  for not too large  $B_0^2$  can be represented in the form

$$\lambda_0 = \frac{1}{T_0} + D_0 B_0^2 - C B_0^4 + \cdots, \tag{45}$$

where  $D_0$  is the coefficient of diffusion of neutrons that have come into complete thermodynamic equilibrium in an unbounded medium (i.e., are Maxwellian), and the coefficient C characterizes the deviation of the spectrum from the Maxwellian one. This coefficient is larger, the greater the value of  $|D_1-D_2|$  and the time required for establishment of the Maxwellian spectrum. Thus, the deviation of the dependence  $\lambda_0(B_0^2)$  from linearity is due to the fact that the spectrum is depleted of neutrons with large values of the diffusion coefficient—there is an effective "cooling" of the spectrum due to the escape of fast neutrons. The effect was therefore called "diffusion cooling," and the coefficient C is the "coefficient of diffusion cooling." These results showed that

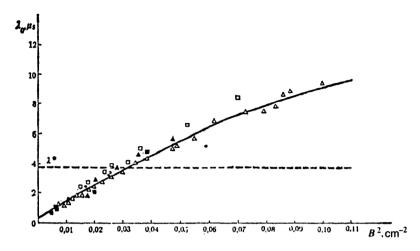


FIG. 5. Dependences of  $\lambda_0$  on  $B_0^2$  according to the measurements of various authors. The solid curve is the approximation of Eq. (45).<sup>26</sup>

measurement of the dependence of  $\lambda_0$  on  $B_0^2$  is an effective method of determining the macroscopic parameters of the interaction of thermal neutrons with matter: Three physical parameters possessing a transparent interpretation can be measured. All this attracted great interest on the part of the scientific community in this method and stimulated its widespread adoption.

# 3. Phenomenological theory of thermalization and diffusion of thermal neutrons with allowance for the velocity distribution

For the correct analysis of the rapidly accumulating experimental information on the diffusion-cooling effect and other effects associated with the deviation of the spectrum of neutrons in a medium from the Maxwellian one, a consistent adequate theory was needed. Such a theory was developed in Refs. 24 and 25. The basic idea of this theory was that the deviation of the neutron spectrum from the Maxwellian distribution  $M_T$  should be represented in the form of an expansion with respect to a complete orthonormal (with weight  $M_T$ ) system of polynomials in the velocity<sup>24</sup> (v representation) or energy<sup>25</sup> (E representation). For example, in the v representation the neutron flux density has the form

$$\Phi_0(\mathbf{r}, v, t) = v M_T(v) \sum_{n=0}^{\infty} \psi_n(\mathbf{r}, t) P_n(v), \qquad (46)$$

where  $P_0=1$ ,  $P_1=v-\langle v\rangle_T$ , etc. Restricting ourselves to the first m terms of this expansion (m-group approximation) and substituting them in (9), we readily obtain a system of equations for the functions  $\psi_n(\mathbf{r},t)$ , in which the coefficients are the matrix elements of transitions between the groups of the quantities  $vl_{tr}(v)$  and  $v'\Sigma_s(v'\rightarrow v)$ :

$$D_{nn'} = \left\langle n \left| \frac{1}{3} v l_{tr}(v) \right| n' \right\rangle, \quad \gamma_{nn'} = \left\langle n \left| v' \Sigma_s(v' \to v) \right| n' \right\rangle.$$

These, in their turn, can be calculated in various models of the matter. In the majority of the cases that are of practical importance, the group expansion converges rapidly. Already the 2-group approximation makes it possible to calculate the coefficient C to an accuracy of 3%. This theory describes, in

the framework of the diffusion approximation and from a unified standpoint, the final stage of moderation—the thermalization of the neutrons and the behavior of thermalized neutrons in both unbounded and bounded media, i.e., it establishes a "bridge" between the age and diffusion theories. By means of this theory, it was possible to explain and predict some important properties of the interaction of thermal neutrons with matter. For example, it follows from this theory that the spectrum of neutrons from a pulsed source in a homogeneous unbounded medium approaches the equilibrium spectrum in accordance with the exponential law

$$\Phi_0(\mathbf{r}, v, t) = v M_T(v) e^{-t/T_0} [\psi_0(\mathbf{r}) + \psi_1(\mathbf{r})(v) - \langle v \rangle_T) e^{-t/\tau_{\text{th}}};$$
(47)

here  $\tau_{\rm th}$  is called the time of neutron thermalization in matter. In the 2-group approximation, it is equal to  $1/\gamma_{00}$ , in good agreement with experiment.

#### 4. Further development of investigations

The possibility of using the developed theory to make quantitative calculations of the parameters of the interaction of thermal neutrons with matter helped to stimulate a large number of new experimental and theoretical studies in this field. In fact, an important new direction in neutron physics arose. International conferences and symposia began to be held on this subject. New interesting effects were discovered. For example, it was shown by means of rigorous transport theory that  $\lambda_0$  cannot exceed the minimum value of the product  $\lambda^* \equiv v \Sigma_{tot}(v)$ , which for crystalline moderators is rather small (for Be it is shown by the dashed curve in Fig. 5). Moreover, more detailed experiments showed that in the cases when  $B_0^2$  corresponds to  $\lambda_0 > \lambda^*$  the damping of the neutron flux in a medium of finite volume occurs nonexponentially. However, this circumstance does not hinder determination of the neutron-diffusion parameters from data obtained with blocks of the medium of larger size.

### 5. Some applications

The establishment of the properties of the interaction of neutrons with matter stimulated wide use of the pulsed neutron method for the solution of various applied problems, in particular for the element and nondestructive structural analysis of samples and geological cores.

The neutron methods of investigating media and samples are based on the detection of fluxes of neutrons that have interacted in matter or on the detection of  $\gamma$  rays produced as a result of these interactions. Comparison of the results then obtained with calculated or reference data makes it possible to determine various neutron-physics parameters (for example the diffusion length in the case of stationary sources and the diffusion coefficient and neutron lifetime in the case of pulsed sources). The detection of  $\gamma$  rays also makes it possible to perform an element analysis of matter. An important advantage of the neutron methods is that the neutrons penetrate deep into the investigated systems (samples), and it is therefore possible to determine the properties of matter without destroying these systems.

Especially informative are methods that use a pulsed source of neutrons, since they make it possible to detect fluxes of neutrons or  $\gamma$  rays as functions of an additional variable—the time that has elapsed since the neutron pulse. In particular, it is possible to choose an optimum time interval in which the investigated properties of the matter are most clearly revealed. In addition, the use of a pulsed source makes it possible to eliminate the background of fast neutrons (by switching on the detector only in the intervals between pulses) or, conversely, separating the effect of the fast neutrons.

Pulsed neutron methods are used both to analyze individual samples and for depth investigations of large volumes of matter. In the analysis of individual samples, the damping constant of the neutron flux in them is measured. By varying the geometry of the sample, one can determine its neutronphysics parameters. The simplest experiment of this type is represented by the measurements described in Sec. 4.2. Great possibilities are opened up by the study of two-region systems, in which one of the regions is the investigated sample and the second is chosen in order to optimize the experiment. For example, if it is necessary to measure the absorption of neutrons in a small volume of matter, it is expedient to place the sample at the center of a much larger volume of a weakly absorbing material. Then the damping constant will depend strongly on the absorption of the neutrons in the sample and weakly on the diffusion coefficient. If it is necessary to determine the diffusion coefficient, the investigated sample should be placed at the position at which the gradient of the neutron density is greatest (for example, at the boundary of a medium with vacuum).

Investigations of matter deep in the interior of a large volume are based on the fact that the dependence of the neutron flux on the time from the instant of the neutron pulse is related to the depth of penetration of the neutrons into the matter: The further the neutrons diffuse, the deeper the layers of matter that influence the neutron fluxes (or the fluxes of  $\gamma$  rays from neutron capture). The maximum number of neutrons that carry information about an object placed at a cer-

tain distance from the source is obtained by using a delay time close to the mean time required for a neutron to reach the object and then the detector. Thus, the effect of an object on the neutron flux is similar to what happens in radar or with an acoustic echo, and accordingly it can be called a "neutron echo" (neutron-gamma "echo"). Therefore, by measuring the time dependence of the neutron flux for one and the same position of the source and detector, it is possible to obtain information about the depth distribution of the properties of a medium. Such a method has found most widespread application in geophysics, in particular, in neutron core analysis. For example, by means of pulsed neutron core analysis it is, as a rule, possible to establish whether a borehole has crossed an oil-bearing or water-bearing layer, since the latter, in contrast to the former, usually strongly absorbs neutrons because of the greater mineralization of wa-

More details about the various applications of the pulsed neutron method can be found, for example, in the monographs of Refs. 26 and 27.

#### **CONCLUSIONS**

In the above, we have attempted to give a brief review of the studies of F. L. Shapiro in the field of the physics of neutron transport in various media and systems. We hope that despite the brevity of this review we have succeeded in showing how important these investigations were at their time and how fruitful they proved to be for the future.

<sup>&</sup>lt;sup>1)</sup>The results of these investigations, including Shapiro's dissertation, were, in accordance with the requirements of the time, kept secret and were first published in 1955 at a session of the USSR Academy of Sciences (Refs. 1–3; see also Ref. 4).

<sup>&</sup>lt;sup>2)</sup>The studies of this series (with references to original publications) are also given in Ref. 4, Part 1.

<sup>&</sup>lt;sup>3)</sup>Here and in all that follows, no limits of integration are given if the integral is extended over the complete range of variation of the variable of integration.

<sup>&</sup>lt;sup>4)</sup>Since this equation has the same form as the equation that describes the diffusion of particles with constant velocity, it is called the "single-velocity" approximation.

<sup>5)</sup>Here we do not consider questions of priority, since under the then existing conditions of secrecy the most important investigations in our country were often made by independent groups, and their authors need not have been informed about the existence of parallel studies, to say nothing of studies made abroad.

<sup>&</sup>lt;sup>6)</sup>In particular, investigations were made of two predictions by I. M. Frank and collaborators of "geometric" effects having a strong influence on the neutron multiplication coefficient: the effect of a gap between the uranium and graphite rods ( $\Delta k \sim +1\%$ ) and the effect of reflection of neutrons by water used to cool the outer surface of the rods ( $\Delta k \sim -1\%$ ).

<sup>&</sup>lt;sup>7)</sup>For a detailed bibliography, see the monograph of Ref. 26.

<sup>&</sup>lt;sup>8)</sup>A similar model was used by von Dardel, <sup>12</sup> who obtained similar results.

<sup>&</sup>lt;sup>1</sup>L. V. Groshev, O. I. Kozinets, L. E. Lazareva et al., "Study of the parameters of uranium-graphite heterogeneous systems by the prism method," in Session of the USSR Academy of Sciences on the Peaceful Uses of Atomic Energy, July 1-5, 1955. Sessions of the Section of Physical and Mathematical Sciences [in Russian] (Izd-vo Akad. Nauk SSSR, 1955), p. 21

<sup>&</sup>lt;sup>2</sup> K. D. Tolstov, F. L. Shapiro, and I. V. Shtranikh, "Mean neutron velocities in different media," *ibid*, p. 108.

 <sup>&</sup>lt;sup>3</sup>B. P. Ad'yasevich, O. I. Kozinets, K. D. Tolstov *et al.*, "Measurement of temperature effects in uranium-graphite subcritical systems," *ibid*, p. 132.
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