

This result is to be compared with the direct differentiation of (16):

$$\mathcal{R}' = \sum_{\lambda} (\gamma_{\lambda}^2)' / (E_{\lambda} - E) - \sum_{\lambda} \gamma_{\lambda}^2 E'_{\lambda} / (E_{\lambda} - E)^2. \quad (\text{B9})$$

Equating those terms in (B9) and (B8) which depend on the energy in the same way yields

$$1 = (4mR^2/\hbar^2) \sum'_{\lambda\mu} \gamma_{\lambda}^2 \gamma_{\mu}^2 / (E_{\mu} - E_{\lambda}) \quad (\text{B10})$$

$$(\gamma_{\lambda}^2)' \equiv R d\gamma_{\lambda}^2 / dR = (2b - 1)\gamma_{\lambda}^2 + (2mR^2/\hbar^2) \times [-\gamma_{\lambda}^4 + 2\gamma_{\lambda}^2(E_{\lambda} - \hat{V}) \sum'_{\mu} \gamma_{\mu}^2 / (E_{\mu} - E_{\lambda})], \quad (\text{B11})$$

$$E'_{\lambda} \equiv R dE_{\lambda} / dR = (2mR^2/\hbar^2)(\hat{V} - E)\gamma_{\lambda}^2. \quad (\text{B12})$$

The sum rule (B10) appears to be a new one and is easily verified explicitly for the case of no potential ( $V = b = 0$ ).

It is interesting to note that in the one-level approximation we can use (B11) to arrive at the

natural boundary condition numbers. Then (B11) may be approximated by

$$(\gamma_{\lambda}^2)' \approx (2b - 1)\gamma_{\lambda}^2 \quad (\text{B13})$$

because the term proportional to  $\gamma_{\lambda}^4$ , in (B11), is small unless  $\gamma_{\lambda}^2$  is an appreciable fraction of the sum rule limit. If we now want the total width to be independent of the matching radius we have

$$\Gamma'_{\lambda} = 0 = 2P'_c \gamma_{\lambda c}^2 + 2P_c (\gamma_{\lambda}^2)'. \quad (\text{B14})$$

Using the definition, (58) of the penetrability (B14) leads at once then to the boundary condition number, (64), required to make the shift function vanish.

The above derivation of  $(\gamma_{\lambda}^2)'$  and  $(E'_{\lambda})$  is not valid if the boundary condition numbers are energy dependent. As we noted in the text, the "natural" boundary condition numbers are usually only moderately dependent on the energy: for most purposes the low-energy limit (the value of  $b_c$  which makes  $S_c$  vanish at zero energy) suffices.

## Recent Developments in the Theory and Technology of Chain Reactors

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**E**UGENE Wigner's sixtieth birthday coincides, within two weeks, with the twentieth anniversary of the first fission chain reaction on December 2, 1942. Wigner's influence on the subsequent development of chain reactors has been extraordinary. There are very few aspects of the current theory that were not outlined by him nor very few of the major technological developments of the last 20 years that cannot be traced to his early ideas.

Wigner's major contributions to chain reactors occurred in three separate periods: First, from 1940 to 1945, during the last three years of which he was head of the theoretical group at The University of Chicago Metallurgical Laboratory; 1946-47, when he was research director of what is now the Oak Ridge National Laboratory; and 1952, when he was a full-time adviser to the Du Pont Company during

the design of the Savannah River heavy water plutonium production reactors.

Chain reactors are heavy engineering devices; they also require, for their design, a certain theoretical and physical sophistication. Wigner's great influence on their development, both at the Metallurgical Laboratory and since, can be traced to the facility with which he could pass back and forth between engineering and physics—from a discussion of the probable distribution of energy levels in  $U^{235}$  to a critical examination of the blueprints of the concrete foundations for the Hanford reactors, or from a group-theoretical argument in transport theory to the design of aluminum fuel elements!

The full force of Wigner's skill and energy was particularly well demonstrated at the Metallurgical Laboratory during the fall of 1942 in the crucial arguments that resulted in the decision to concentrate on water-cooled, rather than on gas-cooled,

\*Operated by Union Carbide Corporation for the U. S. Atomic Energy Commission.

plutonium production reactors at Hanford. Originally, the project had planned to cool the plutonium production reactors with helium. This decision seemed obvious since only natural uranium was available; the infinite multiplication factor would therefore be so close to one that the reactor could hardly tolerate a coolant which absorbed any neutrons, or so it was generally believed at the time. Wigner was convinced, however, that even with fuel consisting of natural uranium sheathed in aluminum, the margin of reactivity was sufficient to allow water as coolant and that, as large-scale plutonium producers, the water-cooled systems were much better than the gas-cooled. Such a conclusion required an unusual combination of theoretical insight and practical engineering judgment. Selling the water-cooled system to the whole project in the face of its prior commitment to gas cooling required a forcefulness and political skill which one could hardly, *a priori*, have associated with the same man who introduced group theory into quantum mechanics. That Wigner was able to set the project on this path probably saved a year in the project timetable, and possibly, eventually shortened the war by a comparable length of time.

#### THEORY OF CHAIN REACTORS

In this short review we shall describe the present status of a few of the parts of the theory and the technology of chain reactors which are particularly associated with Wigner's work. We assume that the reader is generally familiar with the theory of neutron chain reactors. We confine our review of theory to four topics, to each of which Wigner has made major contributions: the spectrum of the Boltzmann equation, neutron thermalization, thermal utilization, and resonance absorption.

#### The Spectrum of the Boltzmann Equation

The linear Boltzmann equation governs both the diffusion of neutrons and the transport of radiation. In the latter context it has been the subject of extensive study by astrophysicists since the early part of this century; much additional study has been given the equation because of its application to the theory and design of chain reactors. It is therefore curious that one of the most central questions concerning the linear Boltzmann equation—its spectrum of eigenvalues and eigenfunctions—was not clarified until the work of Wigner in 1959,<sup>1</sup> and independently,

<sup>1</sup> E. P. Wigner, "Mathematical Problems of Nuclear Reactor Theory," in *Proceedings of the Eleventh Symposium in Applied Mathematics* (American Mathematical Society, Providence, Rhode Island, 1959), Vol. 11, p. 89.

of Case in 1960.<sup>2</sup> Case's considerations were based on results found by Van Kampen for the collisionless Boltzmann equation (Vlasov equation) which enters the theory of plasmas.<sup>3</sup>

To understand the significance of the results of Wigner and Case, we consider the simplest example of the linear Boltzmann equation which describes the transport of monoenergetic neutrons in plane geometry, viz,

$$\mu \frac{d\psi(x,\mu)}{dx} + \psi(x,\mu) = \frac{c}{2} \int_{-1}^{+1} \psi(x,\mu') d\mu'. \quad (1)$$

Here,  $\psi(x,\mu)$  is the density of collisions per sec at  $x$  of neutrons whose velocities make an angle,  $\cos^{-1}\mu$ , with the  $x$  axis. The constant mean free path in the diffusing medium has been taken as the unit of length. All secondaries from either scattering or fission are assumed to be produced isotropically and at the same energy as the primary.  $c$  is the number of such secondaries produced per collision.

In an infinite medium the translation invariance of (1) suggests solutions of the form  $e^{-x/\lambda}\phi_\lambda(\mu)$ . With this assumption (1) becomes

$$(1 - \mu/\lambda)\phi_\lambda(\mu) = \frac{c}{2} \int_{-1}^{+1} \phi_\lambda(\mu') d\mu'. \quad (2)$$

We normalize  $\phi_\lambda(\mu)$  so that the integral on the right of (2) is unity. Then if we integrate over  $\mu$ , we obtain the condition

$$1 = c\lambda \tanh^{-1}(1/\lambda) \quad (3)$$

on  $\lambda$ . This equation has just two real roots,  $\pm \lambda_0$ . It would appear from this simple procedure that  $\phi_{\pm\lambda_0}(\mu)$  are all the solutions of the assumed form, but a hint that this is not so is suggested by the spherical harmonics solution of (1).

Suppose we set

$$\psi(x,\mu) = \sum_{l=0}^{\infty} \frac{2l+1}{2} \psi_l(x) P_l(\mu) \quad (4)$$

where the  $P_l$  are Legendre polynomials. If we substitute (4) into (1) and use some well-known properties of the Legendre polynomials, we obtain the infinite coupled set of equations for the  $\psi_l$ :

$$\frac{l+1}{2l+1} \frac{d\psi_{l+1}(x)}{dx} + \frac{l}{2l+1} \frac{d\psi_{l-1}(x)}{dx} + \psi_l(x) = c\psi_0(x)\delta_{l,0}. \quad (5)$$

<sup>2</sup> K. M. Case, *Ann. Phys.* 9, 1 (1960).

<sup>3</sup> N. G. Van Kampen, *Physica* 21, 949 (1955).

Approximations of various orders to this set can be obtained by setting  $\psi_l(x) = 0$  for  $l > L$ . Again we look for solutions of the form  $\psi_l(x) = g_l e^{-x/\lambda}$ . In any odd order  $L$  we find  $\frac{1}{2}(L + 1)$  pairs of oppositely signed roots  $\pm \lambda$  of the following description: one pair which tends rapidly with increasing  $L$  to the solutions  $\pm \lambda_0$  of (3) and  $\frac{1}{2}(L - 1)$  pairs for which  $|\lambda| < 1$ . To each root  $\lambda$  there corresponds an angular distribution  $\phi_\lambda(\mu)$  determined by the coefficients  $g_l$ . As  $L \rightarrow \infty$  the spectrum of  $\lambda$ 's seems to approach the two isolated values  $\pm \lambda_0$  and a continuous spectrum on the interval  $-1 \leq \lambda \leq 1$ .

Wigner first pointed out that there were solutions of (2) corresponding to just such a continuous spectrum. These solutions are singular and are expressible as Schwartz distributions. In fact, one can deduce from (2)

$$\phi_\lambda(\mu) = \frac{c}{2} P \frac{\lambda}{\lambda - \mu} + \{1 - c\lambda \tanh^{-1} \lambda\} \delta(\lambda - \mu), \tag{6}$$

where the  $P$  denotes the Cauchy principal value and  $\delta$  denotes Dirac's delta function. If  $\lambda$  does not lie on the real axis between  $-1$  and  $+1$ , the delta function disappears since  $-1 \leq \mu \leq +1$ , and the principal value becomes an ordinary value. In such a case  $\lambda$  can only equal  $\pm \lambda_0$ , the two roots of (3), and the corresponding  $\phi_\lambda$  are regular. On the other hand, for every other value of  $\lambda$  on the real axis there is a singular solution  $\phi_\lambda(\mu)$  of the form (6). (The value of the coefficient of the delta function is determined by the requirement of normalization.)

About a year after Wigner's observation, Case, following a method invented by Van Kampen in connection with problems of plasma oscillations, also discovered the continuous spectrum. In addition, he proved an all-important completeness theorem whose content is as follows: If  $-1 < \alpha < \beta < 1$ , the singular solutions  $\phi_\lambda(\mu)$  corresponding to that part of the continuous spectrum  $\alpha \leq \lambda \leq \beta$  are complete for functions  $\psi(\mu)$  on the interval  $\alpha \leq \mu \leq \beta$ ; if  $\alpha = -1$  and/or  $\beta = +1$ , the set  $\phi_\lambda(\mu)$ ,  $\alpha \leq \lambda \leq \beta$ , must be augmented by the regular solution  $\phi_{-\lambda_0}(\mu)$  and/or the regular solution  $\phi_{+\lambda_0}(\mu)$ .

Case's completeness theorem allows the solution of (1) with an arbitrary boundary condition on some plane, say  $x = 0$ , to be written as

$$\psi(x, \mu) = A_{+\phi_{+\lambda_0}}(\mu) e^{-x/\lambda_0} + A_{-\phi_{-\lambda_0}}(\mu) e^{x/\lambda_0} + \int_{-1}^{+1} A(\lambda) \phi_\lambda(\mu) e^{-x/\lambda} d\lambda \tag{7a}$$

with

$$\psi(0, \mu) = A_{+\phi_{+\lambda_0}}(\mu) + A_{-\phi_{-\lambda_0}}(\mu) + \int_{-1}^{+1} A(\lambda) \phi_\lambda(\mu) d\lambda. \tag{7b}$$

In cases where  $\psi(0, \mu)$  is completely specified on the interval  $-1 \leq \mu \leq 1$ , the expansion coefficients  $A_+$ ,  $A_-$ , and  $A(\lambda)$  may be determined by use of an orthogonality relation proved by Case. From (2) and the normalization of  $\phi_\lambda(\mu)$  to unit integral over the interval  $-1 \leq \mu \leq +1$ , Case has shown that

$$\int_{-1}^{+1} \mu \phi_\lambda(\mu) \phi_{\lambda'}(\mu) d\mu = 0, \quad \lambda \neq \lambda' \tag{8a}$$

$$\int_{-1}^{+1} \mu \phi_{\pm\lambda_0}^2(\mu) d\mu = N_{0\pm} = \pm \frac{c}{2} \lambda_0^3 \left( \frac{c}{\lambda_0^2 - 1} - \frac{1}{\lambda_0^2} \right), \tag{8b}$$

$$\int_{-1}^{+1} \mu \phi_\lambda(\mu) \phi_{\lambda'}(\mu) d\mu = N_\lambda \delta(\lambda - \lambda'), \quad -1 \leq \lambda, \lambda' \leq 1 \tag{8c}$$

$$N_\lambda = \lambda \left[ (1 - c\lambda \tanh^{-1} \lambda)^2 + \frac{\pi^2 c^2}{4} \lambda^2 \right]. \tag{8d}$$

As an illustration of these orthonormality relations, let us calculate the collision density  $\psi(x, \mu)$  due to a plane, isotropic source  $\frac{1}{2} \delta(x)$  in an infinite medium.  $\psi(x, \mu)$  now satisfies (1) with the source term  $\frac{1}{2} \delta(x)$  added to the r.h.s. Integration across the plane  $x = 0$  shows that the angular distribution there has a discontinuity given by

$$\psi(0+, \mu) - \psi(0-, \mu) = (2\mu)^{-1}. \tag{9}$$

For  $\psi(x, \mu)$  we choose

$$\psi(x, \mu) = A_{+\phi_{+\lambda_0}}(\mu) e^{-x/\lambda_0} + \int_0^1 A(\lambda) \phi_\lambda(\mu) e^{-x/\lambda} d\lambda, \quad x > 0 \tag{10a}$$

and

$$\psi(x, \mu) = -A_{-\phi_{-\lambda_0}}(\mu) e^{x/\lambda_0} - \int_{-1}^0 A(\lambda) \phi_\lambda(\mu) e^{-x/\lambda} d\lambda, \quad x < 0. \tag{10b}$$

From (9) and (10) it follows that

$$(2\mu)^{-1} = A_{+\phi_{+\lambda_0}}(\mu) + A_{-\phi_{-\lambda_0}}(\mu) + \int_{-1}^{+1} A(\lambda) \phi_\lambda(\mu) d\lambda. \tag{11}$$

Using the orthogonality relations (8) it is easy to find

$$A_\pm = (2N_{0\pm})^{-1} \tag{12a}$$

$$A(\lambda) = (2N_\lambda)^{-1}. \tag{12b}$$

Thus the desired solution is

$$\psi(x, \mu) = \frac{1}{2} \left[ \frac{\phi_{+\lambda_0}(\mu) e^{-x/\lambda_0}}{N_{0+}} + \int_0^1 \frac{\phi_\lambda(\mu) e^{-x/\lambda}}{N_\lambda} d\lambda \right], \quad x > 0 \quad (13a)$$

$$\psi(x, \mu) = -\frac{1}{2} \left[ \frac{\phi_{-\lambda_0}(\mu) e^{x/\lambda_0}}{N_{0-}} + \int_{-1}^0 \frac{\phi_\lambda(\mu) e^{-x/\lambda}}{N_\lambda} d\lambda \right], \quad x < 0. \quad (13b)$$

Often  $\psi(0, \mu)$  is not specified over the whole range  $-1 \leq \mu \leq 1$ , but rather on some smaller interval  $\alpha \leq \mu \leq \beta$ . In such a case the expansion coefficients cannot be found by use of the orthogonality relations. An example of such a situation is furnished by Milne's problem. Here neutrons enter the half-space  $0 \leq x \leq \infty$  at  $x = \infty$  and leave at  $x = 0$ . The collision density  $\psi(x, \mu)$  we seek then satisfies (1) with the further conditions

$$\psi(0, \mu) = 0 \quad \mu \geq 0 \quad (14a)$$

$$\psi(x, \mu) \sim \phi_{-\lambda_0}(\mu) e^{x/\lambda_0}, \quad x \gg 1. \quad (14b)$$

The general solution satisfying (14b) can be written

$$\psi(x, \mu) = \phi_{-\lambda_0}(\mu) e^{x/\lambda_0} + A_{+\phi_{+\lambda_0}}(\mu) e^{-x/\lambda_0} + \int_0^1 A(\lambda) \phi_\lambda(\mu) e^{-x/\lambda} d\lambda; \quad (15)$$

(14a) then gives

$$-\phi_{-\lambda_0}(\mu) = A_{+\phi_{+\lambda_0}}(\mu) + \int_0^1 A(\lambda) \phi_\lambda(\mu) d\lambda, \quad 0 \leq \mu \leq 1. \quad (16)$$

The expansion coefficients are thus determined by the expansion of  $\phi_{-\lambda_0}$  in terms of eigenfunctions with non-negative  $\lambda$  on the interval  $0 \leq \mu \leq 1$ . The relation (16), when written out more fully using the explicit form of the  $\phi_\lambda$ , takes the form of a type of singular integral equation for  $A(\lambda)$  whose solution has been considered at length by Muskhelishvili.<sup>4</sup> The analysis required is exacting and too long to be included here, but Case has considered in detail the various cases which arise in neutron transport problems. In particular, he has calculated the important coefficient  $A_+$  in Milne's problem from which the extrapolation distance can easily be calculated.

In addition to the two problems mentioned above, Case also considered in his original paper the more general problem of a plane collimated source in an infinite medium and the problem of the albedo of a

half-space to a collimated neutron beam. Two other workers, Zelazny and Kuszell,<sup>5</sup> have applied Case's method to the problem of two adjacent half-spaces. These same authors have also generalized the method for two groups of neutrons and discussed the albedo of a half-space and the criticality of a slab on this basis.<sup>6</sup> Another generalization of Case's method to anisotropic scattering has been carried out by Mika<sup>7</sup> who solves Milne's problem and the related problem of two adjacent half-spaces and also finds the collision density due to a plane, collimated source anywhere in a half-space, or on the boundary of two half-spaces. Finally, the continuous spectrum of the Boltzmann equation in spherical geometry has been investigated by Kofink.<sup>8</sup>

The discovery of the continuous spectrum closes a very embarrassing gap in transport theory. With the continuous spectrum and Case's expansion theorems, problems in transport theory can be handled much like the problem of the vibrating string, i.e., by expansion in eigenfunctions. It is too early to say how many hitherto intractable problems will be conquered by application of Case's expansion theorems. In any case, the method, being so transparent in principle, robs transport theory of some of its mathematical complexities and ought to have strong heuristic appeal.

### Thermalization of Neutrons

If an ensemble of neutrons is placed in an infinite, nonabsorbing medium it will after a time reach a state of thermodynamic equilibrium. In this state the ensemble will have a Maxwellian distribution of velocities whose temperature is that of the surrounding medium. If the lifetime of the neutrons against leakage and absorption in a finite, absorbing medium is long compared to the time required to establish

<sup>5</sup> R. Zelazny and A. Kuszell, Polish Academy of Sciences, Institute of Nuclear Research report PAN-219/IX, March 1961 (unpublished).

<sup>6</sup> R. Zelazny and A. Kuszell, Polish Academy of Sciences, Institute of Nuclear Research report PAN-226/IX, May 1961 (unpublished).

<sup>7</sup> J. R. Mika, Nuclear Sci. and Eng. 11, 415 (1961).

<sup>8</sup> W. Kofink, Oak Ridge National Laboratory report ORNL-3216 (November 1961) (unpublished).

<sup>4</sup> N. I. Muskhelishvili, *Singular Integral Equations* (P. Noordhoff N. V., Groningen, Holland, 1953), 2nd ed.

thermodynamic equilibrium, this Maxwellian distribution of velocities will still be an adequate approximation. Nevertheless, the steady-state distribution of velocities even in the presence of weak leakage and absorption will deviate somewhat from the Maxwellian distribution.

If the mechanism of energy exchange between the neutrons and the medium is known, the Boltzmann equation which describes the velocity distribution function may then be solved explicitly. The first calculation of this type was carried out by Wigner and Wilkins.<sup>9</sup> These authors assumed the surrounding medium to be a gas of protons in thermodynamic equilibrium. The spatial extent of the gas was taken to be infinite and the entire space dependence of the problem suppressed. The thermodynamic equilibrium of the neutrons was perturbed by the presence of a  $1/v$  absorber, and steady-state solutions, asymptotic to suitably normalized slowing-down spectra, were sought.

The particular case chosen for study by Wigner and Wilkins, viz., a gas of protons, is a fortunate one; for it the integral equation form which Boltzmann's equation takes can be reduced to a Ricatti differential equation and solved numerically with a minimum of labor. This reduction to a differential equation cannot be carried out for a gas of atoms of atomic weight greater than one, but Wilkins<sup>10</sup> has shown that if the gas is composed of very heavy atoms of mass  $M$ , the integral Boltzmann equation may be consistently approximated to terms of order  $M^{-1}$  by a second-order linear differential equation.

Wilkin's differential equation can be written

$$\frac{2\Sigma_s}{M} \left\{ ET \frac{d^2\phi}{dE^2} + E \frac{d\phi}{dE} + \phi(E) \right\} = \Sigma_a\phi(E) \quad (17)$$

where  $\phi(E)$  is the neutron flux per unit energy interval at  $E$ ,  $T$  is the temperature of the heavy gas times Boltzmann's constant, and  $\Sigma_a$  and  $\Sigma_s$  are the macroscopic absorption and scattering cross sections of the heavy gas, respectively. Equation (17) has been studied exhaustively by several authors.<sup>11</sup> With the term  $-D\nabla^2\phi$  added to the r.h.s. to account for the effects of neutron diffusion, it constitutes a versatile and popular tool for exploring the spatial

variation of the thermal-neutron spectrum in reactors.

### Crystal Effects and Chemical Binding

More realistic calculations of neutron energy spectra in reactors require knowledge of the differential cross section  $d^2\sigma/dE d\Omega$  in energy and angle in the thermal range. In this energy range crystal effects and the effects of chemical binding are important. For this reason, the state of aggregation of the surrounding medium plays a decisive role, and the study of neutron thermalization may be able to shed some light on the properties of solids and liquids.

The study of the scattering of slow neutrons by chemically bound nuclei is greatly simplified by the introduction of Fermi's pseudopotential. According to Fermi, at low energies the scattering of slow neutrons by matter can be calculated in the Born approximation using an interaction potential of the form

$$V(\mathbf{r}) = \sum_k \frac{\hbar^2}{2m_k} 4\pi a_k \delta(\mathbf{r} - \mathbf{r}_k), \quad (18)$$

where  $m_k$  is the reduced mass of the neutron and the struck nucleus,  $a_k$  is the scattering length of the struck nucleus, and  $\mathbf{r}_k$  is its position. The sum runs over all nuclei present.

Even after introduction of the pseudopotential approximation, the calculation of  $d^2\sigma/dE d\Omega$  is a complicated matter. Many such calculations have been carried out with varying degrees of approximation. Five materials of central interest in reactor design have been treated in these calculations: graphite, beryllium, ordinary water, heavy water, and zirconium hydride.

In graphite and beryllium the motions of the individual nuclei are represented as superpositions of quantized simple harmonic oscillations of the crystal lattice (phonons). If the frequency distribution  $f(\omega)$  of these phonons is known, then, in principle,  $d^2\sigma/dE d\Omega$  can be calculated. One commonly used frequency spectrum is that introduced by Debye in the theory of specific heats of crystals, viz., a quadratic dependence of  $f(\omega)$  on  $\omega$  up to a maximum. The Debye spectrum has been used by Singwi and Kothari,<sup>12</sup> by Bhandari,<sup>13</sup> and by Nelkin<sup>14</sup> in studies of Be and BeO crystals. More detailed frequency distributions based on the nature of the crystal lattice and the low-temperature behavior of its specific heat have been

<sup>9</sup> E. P. Wigner and J. E. Wilkins, U. S. Atomic Energy Commission declassified report AECD-2275 (1944) (unpublished).

<sup>10</sup> J. E. Wilkins, Chicago Metallurgical Laboratory report CP-2481 (1944) (unpublished).

<sup>11</sup> H. Hurwitz, Jr., M. S. Nelkin, and G. J. Haetler, *Nuclear Sci. and Eng.* **1**, 280 (1956); E. R. Cohen, *ibid.*, **2**, 227 (1957); L. De Sobrino and M. Clarke, Jr., *ibid.* **10**, 388 (1961).

<sup>12</sup> L. S. Kothari and K. S. Singwi, *J. Nuclear Energy* **5**, 342 (1957).

<sup>13</sup> R. C. Bhandari, *J. Nuclear Energy* **6**, 104 (1957).

<sup>14</sup> M. S. Nelkin, *Nuclear Sci. and Eng.* **2**, 199 (1957).

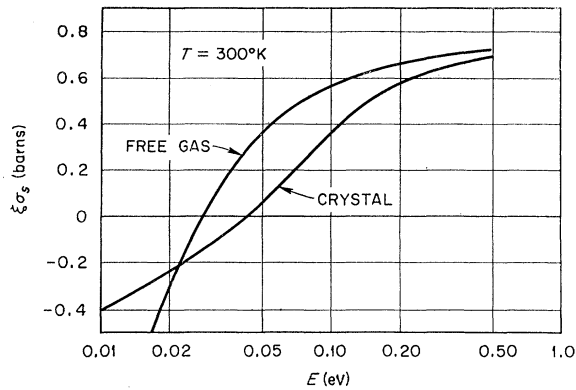


FIG. 1. The theoretical slowing-down power of graphite at 300° K plotted as a function of incident neutron energy and compared with the same quantity for a free gas of carbon atoms [results of D. E. Parks quoted by M. S. Nelkin, "Neutron Thermalization," in *Proceedings of the Eleventh Symposium in Applied Mathematics* (American Mathematical Society, Providence, Rhode Island, 1959), p. 20].

used by Schofield and Hassitt<sup>15</sup> in a calculation of  $d^2\sigma/dE d\Omega$  in graphite. Their results show quantitatively how the crystalline binding of the moderating nuclei slows the thermalization of neutrons in graphite compared to a gas of carbon atoms. This problem was first studied in 1942 by Teller and Metropolis<sup>16</sup> who calculated the rate of heat transfer between a neutron gas at a temperature  $T$  and a graphite lattice at 300°K from the curve of specific heat vs temperature for graphite. Shown in Fig. 1 are the slowing down powers of the crystal atoms and the free atoms at 300°K calculated by D. E. Parks using Schofield and Hassitt's model. For the higher energies the gas is the better moderator; therefore, other things being equal, we expect the thermal-neutron energy spectrum in the crystal to be harder. This expectation is borne out by calculations of the neutron spectrum; shown in Fig. 2, for example, are typical calculated spectra in a mixture of carbon and boron atoms.

The modes of crystalline motion in which large portions of the crystal are coherently excited ("acoustical modes"), such as those described by Debye's frequency distribution, are not the only kinds of motion which may occur. In zirconium hydride, for example, there is another very important mode of motion which can be roughly described as follows: In the lattice of zirconium hydride each proton lies at the center of a tetrahedron of zirconium atoms.

<sup>15</sup> P. Schofield and A. Hassitt, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1959), Vol. 16, p. 217; see also A. Sjolander, *Arkiv Fysik* 14, 315 (1958).

<sup>16</sup> E. Teller and N. Metropolis, Chicago Metallurgical Laboratory Report CP-387 (December, 1942) (unpublished).

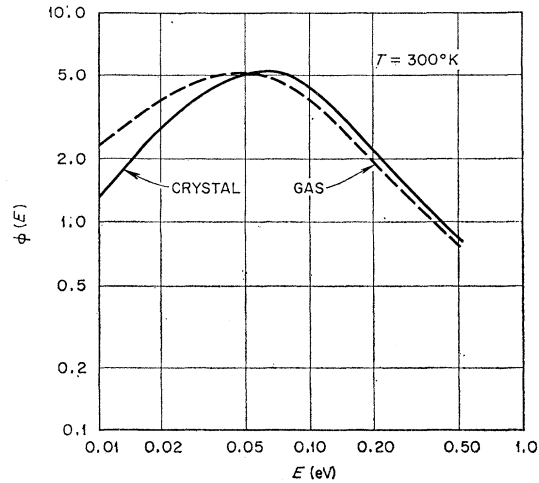


FIG. 2. The flux spectra of neutrons thermalized in mixtures of crystalline graphite and boron and in a free gas of carbon atoms and boron; ratio of carbon atoms to boron = 1890 in both cases [results of D. E. Parks quoted by M. S. Nelkin, "Neutron Thermalization," in *Proceedings of the Eleventh Symposium in Applied Mathematics* (American Mathematical Society, Providence, Rhode Island, 1959), p. 20].

Around this position it is capable of executing simple harmonic oscillations with discrete energies differing by  $\hbar\omega = 0.13$  eV. These "optical" modes of motion will very strongly influence the thermal-neutron spectrum of a reactor containing a lot of zirconium hydride. What happens is this. High-energy neutrons are slowed down into the kinetic energy range from zero to  $\hbar\omega$  with an essentially uniform energy spectrum. Once below  $\hbar\omega$ , these neutrons cannot exchange energy with the zirconium hydride lattice by excitation of the optical modes. Thermalization may proceed by excitation of acoustical modes but this may be slow compared to the processes of leakage and absorption. In any assembly with strong absorption or leakage, therefore, the thermal-neutron spectrum may depart considerably from a Maxwell spectrum. This can be clearly seen in the energy spectra measured by Beyster *et al.*<sup>17</sup> in Fig. 3. The lower spectrum, corresponding to no leakage and 0.49 barns of  $1/v$  absorber per hydrogen atom, is very close to what would be calculated for a free gas of protons and is nearly Maxwellian below about 0.1 eV. The upper curve, corresponding to a relatively light poisoning of 4.95 b per atom, shows a sharp break in the vicinity of 0.13 eV which is due to the strong moderating influence of the first optical level. The departure from a Maxwellian spectrum in this case is large; in cases of equal poisoning in water the

<sup>17</sup> J. R. Beyster, J. L. Wood, W. M. Lopez, and R. B. Walton, *Nuclear Sci. and Eng.* 9, 168 (1961).

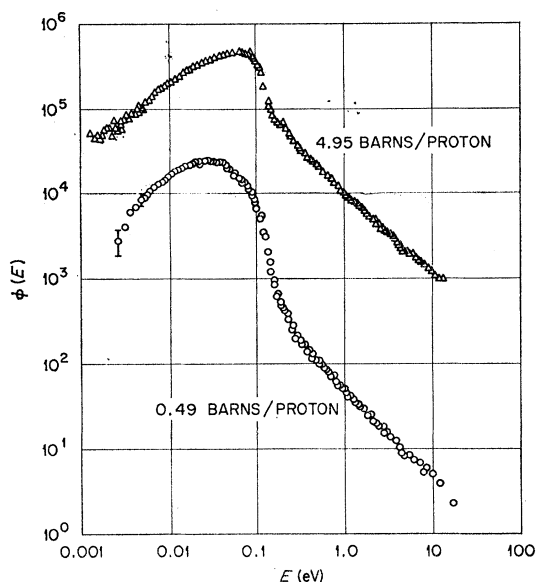


FIG. 3. Flux spectra of neutrons thermalized in zirconium hydride [see reference 17].

spectrum is much less distorted and shows no break at all.

A fraction of the protons in ZrH given by  $\exp(-\hbar\omega/T)$  will be in the first excited state of harmonic motion. These protons may be de-excited by collision with neutrons of energy less than  $\hbar\omega$ , and the neutrons will have their energy increased by  $\hbar\omega$ . If the reactor temperature increases, the proton population of the first excited state of harmonic motion increases, and by energy transfer to the cold neutrons depletes their number. This process can be used to achieve a very high stability against temperature changes; the TRIGA reactor is based on this principle.

#### *Time Behavior; Diffusion Cooling*

Since the early experiments of von Dardel<sup>18</sup> in 1954, it has been recognized that the study of the asymptotic time behavior of a pulse of neutrons injected into a block of material can give important information on the thermalization process. If the block of material can be assigned a unique buckling for thermal neutrons, the asymptotic time behavior of a pulse can be written as  $e^{-\lambda t}$ , where the decay constant  $\lambda$  is given by

$$\lambda = v\Sigma_a + \bar{v}D_0B^2 - CB^4 + \dots \quad (19)$$

$\Sigma_a$  is the macroscopic absorption cross section of the

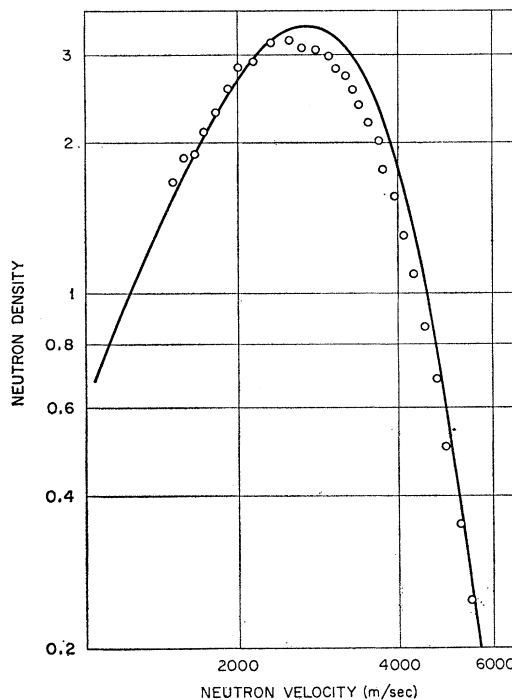


FIG. 4. The asymptotic neutron density spectrum at the center of a  $(5 \text{ cm})^3$ -water cube. Full curve: Maxwellian velocity distribution with moderator temperature [see reference 20].

block, here taken to vary as  $1/v$ ,  $v$  being the neutron velocity,  $\bar{v} = (8T/\pi m)^{1/2}$ ,  $B^2$  is the buckling, and  $D_0$  is the diffusion constant averaged over a Maxwellian flux distribution,  $ET^{-2} \exp(-E/T)$ .

The coefficient  $C$  of the  $B^4$  term is the so-called "diffusion cooling coefficient." The presence of a  $B^4$  term in the expression for  $\lambda$  was first explained by von Dardel and Sjöstrand.<sup>19</sup> They hypothesized a shift of the asymptotic spectrum in a pulsed block toward lower energies due to preferential leakage of high-energy neutrons in a pure Maxwell spectrum. Recently this spectral shift has been observed directly by Beckurts<sup>20</sup> in a small cube of water (see Fig. 4).

Since the diffusion cooling of the Maxwell spectrum competes with the upscattering of the colder neutrons by the moderator, it is to be expected that  $C$  will depend in some way on the energy exchange process between the neutrons and the moderator. Nelkin<sup>21</sup> has shown by a variational technique that  $C$  is related intimately to the reciprocal of  $M_2$ , the second moment of the energy exchange kernel

<sup>19</sup> G. F. von Dardel and N. G. Sjöstrand, *Phys. Rev.* **96**, 1245 (1954).

<sup>20</sup> K. H. Beckurts, *Nuclear Instr. & Methods* **11**, 144 (1961).

<sup>21</sup> M. S. Nelkin, *J. Nuclear Energy* **8**, 48 (1958).

<sup>18</sup> G. F. von Dardel, *Trans. Royal Inst. Technol., Stockholm*, Number 75 (1954); see also, *Phys. Rev.* **94**, 1272 (1954).

$\Sigma_s(E \rightarrow E')$  of neutrons in a Maxwell distribution. Nelkin's relation is

$$C = \frac{(\beta + \frac{1}{2})^2 2\bar{v}D_0^2}{M_2}, \quad (20)$$

where  $\lambda_{tr}(E)$  has been taken proportional to  $E^\beta$  and  $M_2$  is given by

$$M_2 = (T)^{-2} \int_0^\infty dE \int_0^\infty dE' M(E) \times \Sigma_s(E \rightarrow E')(E - E')^2. \quad (21)$$

In water the experimental data on  $C$  are in excellent mutual agreement and yield a value of about  $(4 \pm 1) \times 10^3$  cm<sup>4</sup>/sec;<sup>20</sup> Nelkin has calculated a value of  $(3 \pm 1) \times 10^3$  cm<sup>4</sup>/sec.<sup>22</sup> In crystalline materials the situation is far less satisfactory—not only do various reported experimental values of  $C$  disagree,<sup>20</sup> but the values of  $\lambda$  reported by different experimentalists for the same block of Be differ. Furthermore, some authors have recently shown that reported values of  $\lambda$  sometimes violate rigorous limits that can be placed on them.<sup>23</sup> De Saussure<sup>24</sup> hypothesized that some of these discrepancies might have their origin in a failure to reach an asymptotic time behavior described by a single exponential in the time available for observation of the decaying neutron spectrum. According to him, some of the neutrons are trapped in the high elastic peaks of the transport cross section at energies where the inelastic cross section may be small. In small assemblies, where the over-all leakage probability is comparable to the inelastic scattering probability out of the trap, de Saussure suggests that the instantaneous decay constant may keep decreasing until the last neutron leaves the assembly. Thus, owing to the finite number of neutrons in the initial pulse, an asymptotic spectrum may never be achieved. The trapping of neutrons in the peaks of the transport cross section should also influence the asymptotic energy spectrum in small assemblies, leading to an increased neutron population in the neighborhood of the peaks. This can be seen directly from calculations made by Jha,<sup>25</sup> whose results are displayed in Fig. 5.

<sup>22</sup> M. S. Nelkin, Phys. Rev. 119, 741 (1960).

<sup>23</sup> G. de Saussure, "The Neutron Asymptotic Decay Constant in a Small Crystalline Moderator," presented at the Brookhaven Conference on Neutron Thermalization, April 30–May 2, 1962 (unpublished).

<sup>24</sup> G. de Saussure, Nuclear Sci. and Eng. 12, 433 (1962).

<sup>25</sup> S. S. Jha, J. Nuclear Energy, Pt. A, Reactor Sci. 12, 89 (1960).

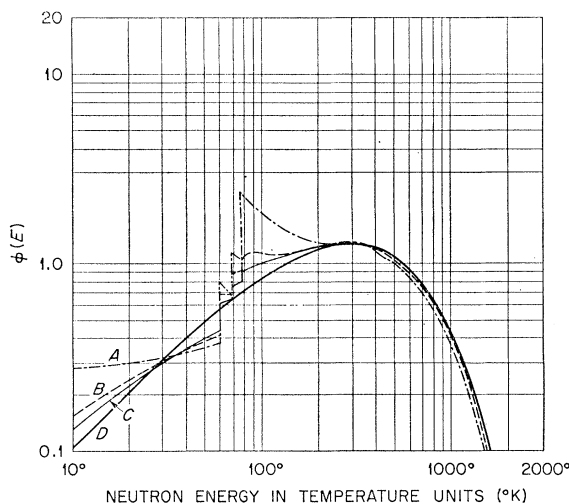


FIG. 5. Flux spectrum in cubes of beryllium of different sizes: A in a 23-cm cube; B in a 35.5-cm cube; C in a 48-cm cube; D in an infinite assembly [see reference 25].

### Thermal Utilization in Lattices

The thermal utilization  $f$  of a reactor is defined as the fraction of all thermal neutrons absorbed which are absorbed in the fuel. Many calculations of  $f$  were done in the design of the first uranium-graphite reactors by Eckart, by Fermi and Weil, by Shockley and Fisk, and by Wigner and Plass. In those early days the thermal utilization was calculated on the following simplified basis:

(1) The transport of neutrons was described by elementary one-velocity diffusion theory, i.e., by the  $P_1$  spherical harmonics approximation.

(2) Spectral shifts in the moderator due to competition between thermalization, leakage, and absorption were ignored.

(3) Thermal-neutron production was taken to be uniform throughout the moderator and zero in the uranium.

(4) The actual lattice cell was replaced by a spherical or cylindrical "Wigner-Seitz" cell of equal volume on whose boundary the normal derivative of the flux vanishes.

Calculations of  $f$  based on approximations (1) to (4) overestimate its true value by the order of 1%. The main errors appear to arise (a) from the use of elementary diffusion theory on the one hand and (b) from neglect of the spectral shift in the moderator on the other. Interestingly, the errors from these two sources have opposite signs, (a) producing an overestimate and (b) an underestimate. Since the total error is positive (a) is the larger of the two sources of error, although (a) and (b) may be of the same order of magnitude.



Wigner and the group at Chicago, and Placzek and the group at Montreal, were the first to try to improve upon the use of diffusion theory by using the  $L = 3$  and  $L = 5$  spherical harmonics approximations to the Boltzmann equation. The resulting calculations are too complicated to be carried out literally, and numerical methods must be applied. Many such numerical calculations have been carried out both by the spherical harmonics method as well as by other methods, e.g., Carlson's  $S_n$  method.<sup>26</sup>

While the numerical solution of higher-order approximations to the Boltzmann equation is a feasible matter, especially in light of the availability of electronic computers today, a method combining the simplicity of elementary diffusion theory with a much enhanced accuracy is desirable. Such a method has been found by Amouyal, Benoist, and Horowitz.<sup>27</sup>

In the method of these authors a variant of one-velocity transport theory is used; otherwise, assumptions (2) to (4) above are still taken as valid. The explicit situation they consider is that of a cylindrical fuel rod of radius  $a$ , surrounded by an annular cavity of thickness  $c - a$  centered in a cylindrical Wigner-Seitz cell of radius  $b$ . They then define three important quantities:  $P_1$ , the probability that neutrons of a uniform, isotropic source in the moderator cross the surface  $r = c$ ;  $\Gamma_1$ , the probability that neutrons entering the moderator at the surface  $r = c$  do not cross that surface again; and  $\Gamma_0$ , the probability that neutrons leaving the moderator at  $r = c$  are captured in the fuel. If we denote by  $J_+$  and  $J_-$ , respectively, the neutron currents crossing the surface  $r = c$  into and out of the moderator, and by  $Q$  the source strength of thermal neutrons in the moderator, a neutron balance over the entire cell then gives

$$Q\pi(b^2 - c^2) = Q\pi(b^2 - c^2)(1 - P_1) + 2\pi cJ_- \Gamma_0 + 2\pi cJ_+ \Gamma_1. \quad (22)$$

A neutron balance over the region  $r < c$  gives

$$2\pi cJ_- \Gamma_0 = 2\pi cJ_- - 2\pi cJ_+. \quad (23)$$

$f$  is defined by

$$f = \frac{2\pi cJ_- \Gamma_0}{Q\pi(b^2 - c^2)}. \quad (24)$$

From these three equations it follows that

$$\frac{1}{f} - 1 = \frac{1}{P_1} \left( 1 - P_1 + \frac{1 - \Gamma_0}{\Gamma_0} \Gamma_1 \right). \quad (25)$$

Amouyal *et al.* now note that the quantities  $\Gamma_1$  and  $\Gamma_0$  depend on the angular distributions of the neutrons crossing the surface  $r = c$  into and out of the moderator, respectively. In general, these angular distributions will depend on the fluxes in the two regions, which are in a sense what is being sought. Amouyal *et al.* assume at this point that both of these angular distributions are the same as the angular distribution of neutrons escaping uncollided across a plane from a semi-infinite, homogeneous, isotropic source, i.e., the angular distribution is proportional to the cosine of the angle the neutron velocity makes with the normal.

With this assumption it is possible to relate  $\Gamma_1$  and  $P_1$  by use of the reciprocity theorem of transport theory<sup>28</sup>:

$$\Gamma_1 = \frac{4V_1}{S_1} \Sigma_{1c} P_1. \quad (26)$$

Here  $V_1$  is the volume of the moderator per unit length,  $S_1$  is the inner surface of the moderator per unit length, and  $\Sigma_{1c}$  is the macroscopic capture cross section of the moderator. An analogous relation holds for  $\Gamma_0$ , viz.,

$$\Gamma_0 = \frac{4V_0}{S_1} \Sigma_{0c} P_0 \quad (27)$$

where  $V_0$  is the fuel volume per unit length,  $\Sigma_{0c}$  the fuel capture cross section, and  $P_0$  the escape (non-absorption) probability of neutrons from a uniform, isotropic source in the fuel.

The greatest error in the use of diffusion theory comes from the fuel region where the dip in the flux is badly underestimated. In the moderator, on the other hand, the flux is rather accurately described by diffusion theory except in a small and unimportant region near the fuel boundary. Amouyal *et al.* therefore prescribe that the calculation of  $P_1$  is to be done with diffusion theory. The boundary condition on the flux at the cell surface is that its normal derivative vanishes. At the inner surface of the moderator,  $r = c$ , the boundary condition is that the linear extrapolation distance  $\lambda$  be that given by Davison and Kushneriuk<sup>29</sup> for the surface of a purely absorb-

<sup>26</sup> B. G. Carlson, Los Alamos Scientific Laboratory report LA-1891 (1955) (unpublished); see also B. G. Carlson and G. I. Bell, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1959), Vol. 16, p. 535.

<sup>27</sup> A. Amouyal, P. Benoist, and J. Horowitz, *J. Nuclear Energy* 6, 79 (1957).

<sup>28</sup> K. M. Case, F. de Hoffmann, and G. Placzek, *Introduction to the Theory of Neutron Diffusion* (U. S. Government Printing Office, Washington, D. C., June 1953), Vol. 1.

<sup>29</sup> B. Davison and S. A. Kushneriuk, Atomic Energy of Canada Report MT-214 (March 30, 1946) (unpublished).

ing cylindrical hole of radius  $c$ . That this boundary condition is correct follows from the fact that  $P_1$  is the probability of *crossing* the surface  $r = c$ .

The result for  $P_1$  is

$$P_1 = \left[ 1 + \kappa^2 b^2 C + \frac{b^2 - c^2}{c} \Sigma_{1c} \frac{3}{2} \lambda \right]^{-1} \quad (28)$$

where  $\kappa$  is the inverse diffusion length in the moderator and

$$C = \frac{1}{2} \left[ \frac{b^2}{b^2 - c^2} \log \left( \frac{b}{c} \right) - \frac{3}{4} + \frac{c^2}{4b^2} \right] \quad (29)$$

if  $\kappa b \ll 1$  as is usual.

Diffusion theory is hopelessly inadequate for calculating  $P_0$ . For this purpose Amouyal *et al.* avail themselves of a method that depends for its success on just those circumstances which invalidate diffusion theory, viz., the fact that the fuel region is small and the fuel strongly absorbing. The method is that of multiple collisions. In its usual form the calculation for  $P_0$  is done by treating the first few collisions exactly and assuming for the remainder that the neutron source after the last collision is spatially uniform. Thus,

$$\begin{aligned} 1 - P_0 &= (1 - \gamma)P^{(1)} + \gamma P^{(1)}(1 - \gamma)P^{(2)} \\ &+ \cdots + \gamma^{N-1}(1 - \gamma) \prod_{i=1}^N P^{(i)} \\ &+ \gamma^N \prod_{i=1}^N P^{(i)} \frac{(1 - \gamma)P^{(1)}}{1 - \gamma P^{(1)}}, \end{aligned} \quad (30)$$

where  $\gamma$  is the fraction of scattering in the fuel,  $P^{(n)}$  is the probability that neutrons from a uniform, isotropic source that have had exactly  $n - 1$  collisions will also have an  $n$ th collision, and  $P^{(n)}$  has been taken equal to  $P^{(1)}$  for  $n > N$ . For isotropic scattering it can be shown that  $P^{(n+1)} \geq P^{(n)}$ , so that (30) gives an overestimate of  $P_0$ .

When  $N = 1$ , (4) reduces to

$$P_0 = \frac{1 - P^{(1)}}{1 - \gamma P^{(1)}} \quad (31)$$

which is a particularly useful formula since  $P^{(1)}$  has been tabulated by Case, Placzek, and de Hoffmann.<sup>28</sup> This equation can, moreover, be derived from a variational solution of the transport equation and is therefore expected to be rather accurate. Comparisons of it with  $N = 2$  calculations done by Amouyal *et al.* and exact calculations done by Stuart<sup>30</sup> bear this out for fuel rods of usual diameters.

Amouyal *et al.* have compared values of  $f$  for

<sup>30</sup> G. W. Stuart, *Nuclear Sci. and Eng.* **2**, 617 (1957).

practical lattices with values found by Carlson's  $S_8$  numerical solution of the Boltzmann equation. These comparisons show that the method of Amouyal *et al.* reduces the error in  $f$  due to transport corrections to the order of one-tenth of one percent, as can be seen from Table I.

TABLE I. Typical values of  $f$  in a uranium-graphite cylindrical lattice according to Amouyal *et al.*<sup>a</sup>

$c$ (cm)	Diffusion theory	$S_8$	Amouyal <i>et al.</i> <sup>b</sup>
1.3	0.8849	0.8683	0.8687
1.6	0.8869	0.8720	0.8718
2.5	0.8901	0.8799	0.8794
3.5	0.8953	0.8877	0.8873

<sup>a</sup> The parameters of the cell used were:  $b = 11.9$  cm,  $a = 1.3$  cm,  $\Sigma_{0c} = 0.3230$  cm<sup>-1</sup>,  $\Sigma_{1c} = 3.118 \times 10^{-4}$  cm<sup>-1</sup>,  $\Sigma_{0t} = 0.7221$  cm<sup>-1</sup>,  $\Sigma_{1t} = 0.3721$  cm<sup>-1</sup>.

<sup>b</sup> See reference 27.

### Resonance Absorption in Lattices

In a pioneering work completed in the second half of 1941 and first published in the open literature in 1955,<sup>31</sup> Wigner demonstrated the calculation of the resonance absorption in bulk uranium and uranium oxide from the scanty data of Fermi and Anderson<sup>32</sup> on the parameters of the predominant resonance lines. So complete were his calculations that they have served as a model for all succeeding work; indeed, the ingenious manner in which he solved the difficult two-region, energy-dependent problem of strict transport theory involved in the calculation of resonance absorption in lattices alone opened the door to further research. This research has largely taken the form of more accurate evaluation of expressions first derived by Professor Wigner and more accurate accounting for effects mentioned by him.

Wigner's basic result is an expression for the effective resonance integral  $I$  of an isolated sphere of absorber in an infinite moderating medium due to *narrow* resonances, i.e., resonances whose extent in lethargy  $\Delta u$  is very much less than  $\xi$ , the logarithmic energy decrement per collision in the absorbing material. His formula for a single resonance and generalized to any shape lump can conveniently be written<sup>33</sup>

<sup>31</sup> E. P. Wigner, E. Creutz, H. Jupnik, and T. Snyder, *J. Appl. Phys.* **26**, 260 (1955).

<sup>32</sup> H. L. Anderson, *Phys. Rev.* **80**, 499 (1950).

<sup>33</sup> See, for example, L. Dresner, *Resonance Absorption in Nuclear Reactors* (Pergamon Press, New York, 1960); or L. W. Nordheim, "The Theory of Resonance Absorption," *Proceedings of the Eleventh Symposium in Applied Mathematics* (American Mathematical Society, Providence, Rhode Island, 1959), Vol. 11, p. 58.

$$\begin{aligned}
 NI = & \int_{\Delta u} \frac{\Sigma_p \Sigma_a(u)}{\Sigma_t(u)} du \\
 & + \int_{\Delta u} \left[ \frac{(\Sigma_t(u) - \Sigma_p) \Sigma_a(u)}{\Sigma_t(u)} \right] \\
 & \times [1 - P^{(1)}(\Sigma_t)] du, \quad (32)
 \end{aligned}$$

where  $N$  is the atomic density of absorber atoms,  $u$  is the lethargy, and  $\Sigma_p$ ,  $\Sigma_a$ , and  $\Sigma_t$  are the potential scattering, absorption, and total macroscopic cross sections of the lump, respectively.

The case of interest to Wigner in 1941 was that of uranium oxide lumps whose linear dimensions were relatively large compared with  $\Sigma_p^{-1} > \Sigma_t^{-1}$ . In the limit of very large dimensions the function  $P^{(1)}(\Sigma_t)$  can be well approximated by

$$1 - S/4V\Sigma_t$$

where  $S/V$  is the surface-to-volume ratio of the absorbing lump. Adoption of this expression for  $P^{(1)}$  will then reduce (32) to a formula for the effective resonance integral in which the first of the two terms is a constant and the second is proportional to the surface-to-mass ratio  $S/M$  of the lump. The first term is the effective resonance integral of an infinite homogeneous medium of the absorber. The values of both these terms can be calculated from the resonance data by integration.

The two-term formula of Wigner was the subject of an interesting discussion at the 1955 Geneva conference. There Gurevich and Pomeranchouk<sup>34</sup> reported work carried out in the U.S.S.R. in 1943 which also led to a two-term formula, this time a constant term and one which was proportional to the square root of the surface-to-mass ratio of the lump. The source of the difference was clarified at the conference by Wigner, who noted that the Russian work took no account at all of neutron moderation by the absorber. This neglect can only be justified for small lumps and in this limit Wigner's more general theory gives the same results as the restricted theory of Gurevich and Pomeranchouk. That this is the case was originally shown by Wigner as follows:

In the limit of large dimensions  $1 - P^{(1)} \approx (\Sigma_t \bar{l})^{-1}$  where  $\bar{l}$  is an abbreviation for  $4V/S$ ; in the limit of very small dimensions  $P^{(1)} \approx 0$ . Wigner notes that by setting  $P^{(1)} \approx \Sigma_t \bar{l} (1 + \Sigma_t \bar{l})^{-1}$  the integrations in (32) can be easily carried out for all lump sizes. Detailed studies have shown the error produced by Wigner's rational approximation to be of the order of 10% or less.<sup>33</sup>

<sup>34</sup> I. I. Gurevich and I. Y. Pomeranchouk, *Proceedings of the International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1956), Vol. 5, p. 466.

In the case of what Gurevich and Pomeranchouk call blocked resonances, the dependence of  $I$  on  $S/M$  is given by

$$\left[ 1 + \left( \frac{m}{4\sigma_p} \right) \frac{S}{M} \right]^{1/2},$$

where  $m$  and  $\sigma_p$  are the prorated mass and potential scattering cross sections, respectively, of the absorber lump per absorber atom (the molecular mass in the case of a pure substance). For sufficiently small  $S/M$ , this expression can be written as a linear expression in  $S/M$ ; for large values of  $S/M$ , on the other hand, it is proportional to  $(S/M)^{1/2}$ . The constant term in the Russian formula comes from unblocked resonances for which (32) gives the proper, size-independent, infinite-dilution limit for  $I$ .

The use of Wigner's rational approximation for  $P^{(1)}$  has another important consequence. With it, (32) can be manipulated into a formal identity with its first term, save that  $\sigma_p = \Sigma_p/N$  must be replaced by  $\sigma_p + (N\bar{l})^{-1}$ . From this fact two very interesting and highly practical, though approximate, equivalence results may be derived.<sup>35</sup> The first of these is the following: Let us consider lumps containing a particular absorber and various amounts of other moderating materials. Then all such lumps with the same value of  $\sigma_p + (N\bar{l})^{-1}$  have the same effective resonance integral  $I$ . The second equivalence result states that the effective resonance integral for a lump is the same as that for a homogeneous mixture of moderator and the same absorber in which the potential scattering cross section per absorber atom is  $\sigma_p + (N\bar{l})^{-1}$ . Both of these theorems have been checked against experiment and agreement is excellent.<sup>33,35</sup>

Not all resonances are narrow with respect to collisions in their own material, though virtually all are narrow with respect to collisions in the common moderators. This was recognized by Wigner, who chose some simple approximations adequate for the purpose of estimating resonance absorption in uranium oxide. The situation inspired later authors to consider the problem of resonance absorption by a mixture of an absorber for which  $\xi_a \ll \Delta u$  and a moderator for which  $\xi_m \gg \Delta u$ . Those considerations were usually based on the idealization of assuming  $\xi_a = 0$ , i.e., assuming the mass of the absorber nucleus to be infinite. A formula similar in form to (32) can be derived by an argument rather similar in style to that originally used by Wigner for narrow resonances,

<sup>35</sup> E. Hellstrand, *J. Appl. Phys.* **28**, 1493 (1957); E. Hellstrand, P. Blomberg, and S. Horner, *Nuclear Sci. and Eng.* **8**, 497 (1960).

where the key point was to divide the neutrons into two classes, depending on whether their last collisions were with an absorber atom or with a moderator atom. This formula is

$$NI = \int_{\Delta u} \frac{\Sigma_a(u)\Sigma_m}{\Sigma_a(u) + \Sigma_m} du + \int_{\Delta u} \frac{\Sigma_a^2(u)}{\Sigma_a(u) + \Sigma_m} P_0(\Sigma_t) du, \quad (33)$$

where  $P_0$  is the escape (nonabsorption) probability of neutrons from a uniform, isotropic source in the fuel.

Adler, Hinman, and Nordheim and Spinrad, Chernick, and Corngold have suggested the use of (31) for  $P_0$ . When Wigner's rational approximation to  $P^{(1)}$  is combined with the flat-flux approximation (31) to  $P_0$  (33) is also reduced to a formal identity with the first term of (32), except in this case  $\sigma_r$  must be replaced by  $\sigma_m + (Nl)^{-1}$ .

In the actual evaluation of the integrals in (32) and (33), the broadening of the resonance lines due to the thermal motion of the absorber nuclei must be taken into account. In all practical calculations the thermal motion of the absorber nuclei is described by a Maxwell distribution of velocities. The actual shape of the resonance line is then a convolution of a Gaussian function and the natural line shape, sometimes called the Voigt profile.

In the theory of resonance absorption a variety of integrals of functions of Voigt's profile arise. The first of these integrals to be studied in the years following Wigner's original work was the first term of (32), which is characteristic of absorption in homogeneous systems as well as in heterogeneous systems if the rational approximation to  $P^{(1)}$  is used. The tabulation of this integral, first done in 1956,<sup>36</sup> made possible the accurate calculation of the temperature variation of effective resonance integrals. In 1958 Adler, Hinman, and Nordheim<sup>37</sup> tabulated the second term of (32) for slabs, cylinders, and spheres, thus avoiding use of the rational approximation for narrow resonances in heterogeneous media. If scattering by the absorber is neglected, the second term of (32) and the second term of (33) are formally identical so that the tabulation of Adler *et al.* can also be used to treat wide resonances.

Wigner's original calculation of the effective resonance integrals of uranium metal and oxide was

based on the early resonance data of Fermi and Anderson. New experiments were reported starting around 1955, and interest in the recalculation of the resonance integrals was reawakened. An early attempt was made by Dresner,<sup>36</sup> who in 1956 calculated the resonance integrals of uranium and thorium in homogeneous mixtures in the narrow resonance approximation including the Doppler effect. In 1958 three calculations of resonance integrals in heterogeneous media by Dresner,<sup>38</sup> by Chernick and Vernon,<sup>39</sup> and by Adler, Hinman, and Nordheim<sup>37</sup> were published. All three were similar in rationale, the main feature being the use either of Wigner's narrow resonance formula (32) or the infinite mass absorber formula (33) depending on whether the energy loss on scattering was large or small compared with the width of the resonance. In 1960 a similar calculation was reported by Rothenstein.<sup>40</sup> These four calculations represent the quintessence of Wigner's method; for, while refined in various regards (e.g., inclusion of the Doppler effect through exact evaluation of various integrals involving Voigt's profile, avoidance by Adler *et al.* of the rational approximation to  $P^{(1)}$ , allowance for the Porter-Thomas<sup>41</sup> fluctuation in the neutron widths in estimating the contribution of unresolved resonances, inclusion of contributions from  $p$ -wave resonances), they all depend on the use of Wigner's ingenious ideas for solving the energy-dependent transport problem involved in resonance absorption. Moreover, all give quite good agreement with direct measurements of the effective resonance integrals of uranium and uranium oxide (agreement in the case of thorium and thorium oxide is not as good as for uranium and uranium oxide, but this appears to be due at least in part to inaccuracies in the resonance data and the influence of negative energy resonances on the  $1/v$  contribution).<sup>42</sup>

Most of the effective resonance integral in uranium lumps can be attributed to resonances for which the choice of the narrow resonance or infinite mass absorber approximation is unambiguous. Nevertheless, there are resonances for which neither extreme approximation is applicable. To deal with these intermediate resonances, Nordheim<sup>42</sup> has suggested the following procedure: The resonance is assumed narrow to *moderator* collisions. If neutrons make their

<sup>36</sup> L. Dresner, Nuclear Sci. and Eng. 1, 68 (1956).

<sup>37</sup> F. T. Adler, G. W. Hinman, and L. W. Nordheim, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy* (United Nations, New York, 1959), Vol. 16, p. 142.

<sup>38</sup> L. Dresner, Oak Ridge National Laboratory Report ORNL-2659 (February 1959) (unpublished).

<sup>39</sup> J. Chernick and R. Vernon, Nuclear Sci. and Eng. 4, 649 (1958).

<sup>40</sup> W. Rothenstein, Nuclear Sci. and Eng. 7, 162 (1960).

<sup>41</sup> C. E. Porter and R. G. Thomas, Phys. Rev. 104, 483 (1956).

<sup>42</sup> L. W. Nordheim, Nuclear Sci. and Eng. 12, 457 (1962).

last collision in either the moderator or the fuel, the probability they will make their next collision in the same region is taken to be the first-collision probability from a uniform isotropic source in that medium,  $P^{(1)}$ . With these assumptions, the collision density *per unit energy* in the lump,  $F(u)$ , satisfies the equation

$$F(u) = P^{(1)}[\Sigma_i(u)] \int_{u-\epsilon}^u \frac{F(u') \Sigma_s(u')}{1 - \alpha \frac{\Sigma_s(u')}{\Sigma_i(u')}} du' + \{1 - P^{(1)}[\Sigma_i(u)]\} \Sigma_i(u) \frac{e^{-u}}{E_0}, \quad (34)$$

where all quantities refer to the lump,  $\alpha = (A - 1)^2 / (A + 1)^2$ ,  $A$  being the absorber mass number,  $\epsilon = \ln(1/\alpha)$ , and  $E_0$  corresponds to the zero of lethargy.

This single integral equation is now integrated numerically; Nordheim and Kuncir<sup>42</sup> have written a flexible and rapid program for the IBM-7090 for doing this.

Presented in Table II are the results of Nordheim's calculations of the effective resonance integrals of

TABLE II. Comparison of Nordheim's calculated values for the resonance integrals of  $U^{238}$  and their temperature coefficients at 300° K with Hellstrand's empirical formulas.

		Uranium metal (density 18.7 g/cm <sup>3</sup> )				
Rod radius (cm)		0.1055	0.211	0.422	0.844	1.69
$S/M$ (cm <sup>2</sup> g <sup>-1</sup> )		1.013	0.507	0.254	0.127	0.0634
Calculated Resonance Integral (barns) <sup>a</sup>		28.95	21.41	16.01	12.19	9.48
$2.95 + 25.8 (S/M)^{1/2}$ (barns) <sup>b</sup>		28.91	21.28	15.95	12.18	9.45
$\beta \times 10^{2c}$		0.98	0.75	0.63	0.56	0.57
		UO <sub>2</sub> (density 10.2 g/cm <sup>3</sup> )				
Rod radius (cm)		0.125	0.25	0.50	1.0	2.0
$S/M$ (cm <sup>2</sup> g <sup>-1</sup> )		1.570	0.785	0.393	0.196	0.098
Calculated Resonance Integral (barns) <sup>a</sup>		38.23	27.74	21.05	16.32	12.76
$4.45 + 26.6 (S/M)^{1/2}$ (barns) <sup>b</sup>		37.76	27.95	21.10	16.23	12.79
$\beta \times 10^{2c}$		1.26	1.00	0.79	0.67	0.55
Rod radius (cm)		0.4		0.85		1.4
$\beta \times 10^2$ Calculated <sup>a</sup>	for U metal <sup>e</sup>	0.64		...		0.55
Observed <sup>d</sup>		$0.64 \pm 0.05$		...		$0.55 \pm 0.05$
$\beta \times 10^2$ Calculated <sup>a</sup>	for UO <sub>2</sub> <sup>c</sup>	0.84		0.69		0.63
Observed <sup>d</sup>		$0.82 \pm 0.06$		$0.69 \pm 0.05$		$0.65 \pm 0.05$

<sup>a</sup> L. W. Nordheim, Nuclear Sci. and Eng. **12**, 457 (1962).

<sup>b</sup> E. Hellstrand, J. Appl. Phys. **28**, 1493 (1957). The experimental values given in the table for uranium metal are 4.5% higher than in this reference, as suggested in a private communication from E. Hellstrand to L. Nordheim noted in reference a.

<sup>c</sup> The temperature dependence of the effective resonance integral is represented by the formula  $I(T) = I(300) [1 + \beta (T^{1/2} - 17.3)]$ ,  $T$  in °K.

<sup>d</sup> E. Hellstrand, P. Blomberg, and S. Horner, Nuclear Sci. and Eng. **8**, 497 (1960).

uranium and uranium oxide rods and, for comparison, Hellstrand's direct measurements.<sup>35</sup> By assuming an entirely reasonable contribution of 1.7 b for resonances above 30 keV and  $p$ -wave resonances, perfect agreement is achieved, supporting to the fullest Nordheim's contention that "the goal of a reliable method for calculation of resonance integrals from resonance data has been achieved."

#### TECHNOLOGY OF CHAIN REACTORS

Wigner has contributed directly to the development of research reactors, of plutonium production reactors, and of power reactors. Above all, he has done much to clarify the grand strategy of nuclear reactor development. We therefore begin our short summary of reactor technology with an account of the "philosophy" of reactor development; special reactor types with which Wigner has been associated will be discussed later.

#### The Motivation for Reactor Development

Perhaps the greatest difficulty in reducing nuclear reactor technology to large-scale practice is not technical; it is rather a question of giving a fully convincing motivation for the development. The motivation is clear for those parts of the technology where reactors can do something unique. Nuclear power in a submarine needs no further justification; there is no other prime mover that can operate for very long times at high power without a supply of oxygen.

Nuclear reactors for civilian power are a very different matter. If nuclear electricity had turned out to be very much cheaper than conventional electricity, the motivation for the development of nuclear power would have been straightforward. As it is, nuclear electricity is, if anything, more expensive than conventional power; moreover, the relative economics of the two energy sources depends very strongly on the local situation. Thus, consider the recently announced 325-MW boiling water reactor to be built for Pacific Gas and Electric Company at Bodega Bay, California; this plant represents the current state of the American art. The estimated cost of electricity from this reactor is 7.4 mills/kWh at 60% load factor, and 5.5 mills/kWh at 90% load factor.<sup>43</sup> A conventional oil-fired plant of the same size built at Bodega Bay would produce electricity at 7.4 mills/kWh at 60% load factor and 5.9 mills/kWh at 90% load factor. According to these estimates, nuclear power at Bodega Bay is economically competitive if the load factor is 60% or

<sup>43</sup> Nucleonics **20**, 26 (1962).

higher. The situation is much more complicated than this, however. In the first place, the cost of fossil fuel at Bodega Bay,  $38¢/10^6$  Btu, is some  $12¢/10^6$  Btu higher than the average cost of fossil fuel in the rest of the United States; a reduction of  $12¢/10^6$  Btu would reduce the cost of electricity from the oil-fired plant by more than 1 mill/kWh. Secondly, if the plant were built in the United Kingdom or in Canada, where utilities are publicly owned and economic custom places the fixed charges on capital investment at 7% instead of 14%, power from the nuclear plant would be cheaper than from the conventional plant by about  $\frac{1}{2}$  mill/kWh. Finally, and perhaps most importantly, the computation of nuclear fuel charges depends very sensitively upon the cost of  $U^{235}$  (Bodega Bay uses  $UO_2$  enriched to 2% in  $U^{235}$ ), the buy-back price for the plutonium in the spent fuel elements, the lease-charge for  $U^{235}$  held up in the reactor, and the price for reprocessing spent fuel elements. These prices are set largely by government fiat; the current prices represent a substantial government subsidy for the Bodega Bay plant. If, for example, private utilities had to pay for nuclear fuel as a normal working capital investment at 12% rather than lease it at the  $4\frac{3}{4}\%$  now prevailing, the cost of power from Bodega Bay would increase by about 0.5 mill/kWh. Leasing nuclear fuel from a privately financed stockpile might reduce the rate to about 7%, in which case the added cost would be 0.2 mill/kWh.

The above example illustrates how fragile is the criterion of economic advantage as a justification for nuclear power. Even in high-cost conventional fuel areas, the balance is close; in low-cost areas, conventional systems have a clear-cut advantage of perhaps 1 mill/kWh over nuclear systems. For this reason some writers, notably Professor Wigner, have argued that the *primary* aim of nuclear energy development ought to be to produce electricity economically for the much longer period of human history when we have run out of high-grade fuels, rather than to make electricity which is marginally competitive when we have a great abundance of fossil fuel.

The energy content of the  $U^{235}$  in the really high-grade uranium ores in the United States (those costing up to \$10/lb of  $U_3O_8$ ) is estimated to be  $0.2 \times 10^{18}$  Btu; this is very small compared to the estimated energy content,  $12 \times 10^{18}$  Btu, in the reserves of coal available in the United States at costs up to  $35¢/10^6$  Btu. In this sense, developing reactors that can economically burn only the  $U^{235}$  in high-grade uranium ores, is not a terribly important task. On the

other hand, the energy content of the uranium and thorium in all of the earth's rocks (at an average concentration of 10 ppm for thorium, 2.5 ppm for uranium) is of the same order as the energy content of the deuterium in the sea—enough to last forever. As the price of uranium increases from \$10/lb to the price of residual uranium, the amount available increases, possibly as the square of the price per pound. The development of reactors that can economically utilize these more expensive ores and therefore provide an inexhaustible energy source would appear to be one of mankind's most important ultimate tasks.

To utilize expensive ores economically, reactors must burn much more than the original  $U^{235}$ . For example, a reactor having a conversion ratio (fissile atoms produced per fissile atom burned) of 0.98, can burn 50 times as much  $U^{238}$  as the original  $U^{235}$  it burns. One can therefore afford burnup charges for the raw material in such a reactor 50 times as high as one can pay in a reactor that burns pure  $U^{235}$ . There is perhaps 2500 times as much uranium at 50 times the present cost as there is at the present cost. Hence reactors with a conversion ratio of 0.98 would increase the economically available energy supply (insofar as the cost is determined by the burnup cost) not merely by a factor of 50, but by the product of factors  $50 \times 2500 = 1.25 \times 10^5$ . Thus, in principle, high-conversion ratio reactors—preferably breeders—increase the economically available supply of energy not only by the isotopic ratio of  $U^{238}$  to  $U^{235}$  (which is 140) but by the product of 140 and the much greater ratio ( $\sim 10^7$ ) of the amount of low-grade ores to the amount of high-grade ore.

This elementary point was realized at the Metallurgical Laboratory; however, in the following decade, during which the direction of the nuclear energy enterprise was determined, people lost sight of the crucial importance of high-conversion ratio reactors. Breeders were generally believed to increase the economically retrievable energy by a factor of 140, not by the much greater factor of  $140 \times 10^7$ . The rediscovery of this point is largely due to the work of Harrison Brown; since Brown's studies, the breeder reactor has occupied an increasingly important position in nuclear technology.

Assuming that breeders themselves are feasible, two auxiliary questions must be settled before one can say with confidence that breeder reactors can serve as the basis for a truly long-term energy system. First, can we place reliable upper limits on the cost of uranium and thorium from low-grade ores; and second, do we see ways of permanently and safely disposing of the radioactive wastes? Of the two ques-

tions, the former is the easier: Work, for example, by Brown *et al.*<sup>44</sup> has shown that thorium plus uranium can be extracted from Conway granites which have an average concentration of  $56 \pm 6$  ppm thorium plus about 10 ppm uranium for costs less than \$80/lb. The amount of thorium in this single deposit is estimated to be at least 25 million tons and possibly more.

The waste disposal problem is very much more complicated, as much as anything, because there is no one kind of liquid radioactive waste. Reactor development proceeds along many parallel paths, and the chemical characteristics of the wastes from one line may not be very much like the chemical characteristics of the wastes from another line. Two divergent views toward the waste disposal question are discernible: one school says dilute and disperse, the other, concentrate and isolate. Among the many methods based on the dilute and disperse philosophy we mention two here: hydrofracture in bedded shale and direct disposal in salt. In the hydrofracture method,<sup>45</sup> one pumps, under pressure, a slurry of grout, containing the radioactivity to be disposed of, into a steel-cased bore hole which has a lateral opening well below the ground water table—perhaps 1000 feet below the surface. The liquid, being under very high pressure, intrudes between planes of bedded rock and forms a solid sheet of concrete perhaps 5 millimeters in thickness and extending over several acres. Since the sheet is solid, and since no liquid can get to it, the radioactivity ought to be permanently immobilized well below the water table. Hydrofracturing experiments to date have behaved fairly well, though two unexpected difficulties have shown up: first, the cement set up too quickly and the sheet did not extend as far as was expected; and second, the grout encountered a vertical fault plane which it invaded to a height some hundreds of feet above the level of the rest of the sheet.

Disposal in salt domes is *a priori* attractive since a cavity dissolved out by water is impervious; enough salt dissolves to saturate the solution after which no more salt dissolves and the water is trapped. Actually, the disposal of liquid radioactive wastes into such formations is made much more difficult because the liquid wastes are very acid; this acid solution in contact with salt generates gas containing oxygen-chlorine compounds which are explosive. On the other hand, salt mines, because of their dryness and

isolation, seem to be attractive vaults for permanent disposal of radioactive materials which are first immobilized. Much of the work on waste disposal is therefore now centered on finding ways to permanently immobilize the wastes—generally in the form of a highly calcined oxide or as a glass. Considerable success has been achieved in calcining liquid wastes in stainless steel pots<sup>46</sup>; these pots, containing solidified radioactive wastes, would then be stored permanently in unused salt mines.

### Power Reactors

In spite of the great ultimate significance of breeder reactors, most of the power reactor development in the United States, and indeed in the whole world, is aimed at the short-range goal of producing nuclear electricity that is economical where conventional fuels are expensive or where fixed charges are low. The main American reactor type, the pressurized-water reactor and its variant, the boiling-water reactor, borrows very heavily from the technology of the Materials Testing Reactor (MTR). The power plant for the Nautilus is essentially a pressurized MTR with zirconium replacing aluminum as the fuel element matrix.

Pressurized-water reactors using highly enriched uranium have proved to be extremely reliable naval power plants. However, as central station plants they run into economic difficulties because highly enriched uranium is so expensive—\$12.00/gram. If  $U^{235}$  is burned in a pressurized-water reactor that operates at a thermal efficiency of 25%, the burnup cost alone would be about 2.40 mills/kWh. The total fuel cost would be perhaps twice this if one takes into account the cost of refabricating and reprocessing partially spent fuel elements. These costs are much too high for central power stations.

On the other hand, the  $U^{235}$  in natural uranium costs about \$1.50/gram. If natural uranium could be burned completely in a pressurized-water reactor, the burnup cost would come to only 0.3 mill/kWh; even if fabrication and fuel inventory costs are included, the fuel cycle cost would remain around 1.5 mills/kWh. At this fuel cycle cost, nuclear power would in many areas be competitive with conventional sources.

Natural uranium in light water will not sustain a high-powered chain reaction because of the relatively large absorption cross section of hydrogen. Thus, in the pressurized-water systems, one must use uranium with at least some enrichment. In general, one tries

<sup>44</sup> M. Gerrard, editor, Oak Ridge National Laboratory Report ORNL-3314 (September 1962) (unpublished).

<sup>45</sup> E. G. Struxness, Oak Ridge National Laboratory Report ORNL-TM-133, (March, 1962) (unpublished).

<sup>46</sup> H. W. Godbee and J. T. Roberts, Oak Ridge National Laboratory Report ORNL-2986 (August 1961) (unpublished).

to design so as to minimize the enrichment; this not only reduces the cost of the initial load of  $U^{235}$ , but it also prolongs the life of the fuel since the lower the enrichment the greater is the fraction of  $U^{238}$  converted into fissile  $Pu^{239}$ . Two different schemes for arranging the enriched fuel are possible: First, highly enriched  $U^{235}$ -bearing plates, comprising a "seed," can be surrounded by a "blanket" of natural or depleted uranium. Plutonium is produced in the blanket and, as the reactor operates, the power maximum shifts from the seed into the blanket; this is the configuration of the Shippingport reactor which is now operating with its third seed and second blanket. Second, one can distribute slightly enriched uranium more or less uniformly throughout the reactor and allow the plutonium to be produced everywhere, not just in the blanket. This latter arrangement, being simpler, is the preferred one for most pressurized-water reactors aimed at producing economical power.

Since metallic uranium is incompatible with very hot water, ceramic materials containing uranium are now used as fuels. The most important of these is  $UO_2$ ; it would not be much of an exaggeration to say that the central technological question in most current power reactor development is the behavior of uranium oxide under the conditions of temperature and radiation encountered in a chain reactor. Fortunately, it has been found that  $UO_2$ , as long as it is close to its stoichiometric constitution, sustains long burnup and high temperature very well. The success of the  $UO_2$ -fueled, boiling-water reactor type can be judged by the previously mentioned cost estimates for the Bodega Bay reactor.

Power reactor technology aimed at achieving short-term, economically competitive, central station power comprises much more than pressurized-water reactors; there are the heavy-water systems, graphite-sodium systems, organic-cooled and moderated systems, and most important, the gas-cooled, graphite-moderated systems. Gas-cooled reactors have been largely ignored in the United States until quite recently, though they have constituted the primary path in many foreign countries, especially in Great Britain and France. The United States' position can be traced to the early decision taken at the Metallurgical Laboratory to concentrate on water cooling rather than on gas cooling. The basis for this original decision was that gas-cooled reactors, being low-power-density devices, are bulkier and require more uranium than do water-cooled reactors. On the other hand, since the neutron economy of the gas-cooled systems is superior, the fuel cycle costs of a gas-cooled reactor should generally be below those of a water-

cooled reactor. The relative advantages of gas cooling and water cooling therefore depend upon the book-keeping. Where fixed charges on capital investment are high, as in the United States, the smaller and cheaper water systems are advantageous; where fixed charges are low, as in the United Kingdom, the larger and more expensive gas-cooled systems with compensating lower fuel costs are attractive.

Even in the United Kingdom, the optimum may not lie with the low-rated, natural-uranium, expensive-to-build Calder Hall-type reactor (so-called Magnox stations because the fuel is sheathed in an Mg-Be alloy called Magnox). By enriching slightly, one can greatly increase both the fuel rating and temperature and decrease the size and capital cost of the reactor. For this reason, the British are now constructing the AGR (advanced gas-cooled reactor) which uses 2.5% enriched  $UO_2$  sheathed in stainless steel. The fuel is rated at 7.7 kW/kg as compared to 5.9 kW/kg in the most modern Magnox stations (Sizewell). The estimated capital cost of large AGR's is \$227/kW as compared with \$286/kW for Sizewell; the power costs in the two cases, however, are estimated to be 8.9 mills/kWh for AGR, 7 mills/kWh for Sizewell, though the cost for AGR ought to go down as  $U^{235}$  becomes cheaper. It is noteworthy that in the United States, gas cooling has been revived and a reactor very much like the AGR is now under construction at Oak Ridge.

The AGR-type reactors use  $UO_2$  fuel elements. Thus, the two dominant lines of power reactor development—pressurized water and gas cooling—which developed quite separately following the original decision to concentrate on water cooling at the Metallurgical Laboratory, now converge, at least as far as their central technical problem, the fuel element, is concerned. Sintered  $UO_2$  is the fuel of choice in both systems.

### Power Breeders

We now turn briefly to breeders and near-breeders, reactors whose development would enable us to tap the inexhaustible residual uranium and thorium in the earth's crust. Breeders are feasible in principle only if  $\eta$ , the number of neutrons emitted per neutron absorbed in the fissile nucleus, exceeds 2. At high energy ( $\sim 1$  MeV and above),  $\eta$  is comfortably above 2 for both  $U^{233}$  and  $Pu^{239}$ , first because radiative capture competes poorly with inelastic scattering, and second because the number of neutrons per fission increases with energy (about 1/8 neutron per MeV). At thermal energy, the lowest lying resonance in  $U^{233}$  happens to have a relatively small radiation



width, in  $\text{Pu}^{239}$  a much higher radiation width. As a result, in  $\text{U}^{233}$   $\eta$  at 2200 m/sec is 2.29, in Pu  $\eta$  is 2.14. Until the advent of the pulsed Van de Graaff, there were almost no reliable data on  $\eta$  in the intermediate energy region. Fortunately this region has now been investigated by Diven and Hopkins<sup>47</sup> and Macklin *et al.*<sup>48</sup> Current values of  $\eta$  in the kilovolt region are given in Table III. The values of  $\eta$  given

TABLE III. Fission neutron yield parameters for  $\text{U}^{233}$  and  $\text{Pu}^{239}$ .  $\nu$  = number of neutrons per fission;  $\alpha = \sigma_c/\sigma_f$ ;  $\eta = \nu/(1 + \alpha)$ .

$E$	$\nu$	$\text{U}^{233}$		$\text{Pu}^{239}$		
		$\alpha$	$\eta$	$\nu$	$\alpha$	$\eta$
0.025 eV	2.51 <sup>a</sup>	0.0932	2.296 <sup>d</sup>	2.89 <sup>a</sup>	0.349	2.143 <sup>e</sup>
30 keV	2.51 <sup>b</sup>	0.109 <sup>c</sup>	2.26	2.89 <sup>b</sup>	0.343 <sup>c</sup>	2.15
175 keV	2.53 <sup>b</sup>	0.098 <sup>c</sup>	2.30	2.91 <sup>b</sup>	0.142 <sup>c</sup>	2.55
400 keV	2.56 <sup>b</sup>	0.078 <sup>c</sup>	2.37	2.94 <sup>b</sup>	0.089 <sup>c</sup>	2.70
1 MeV	2.64 <sup>b</sup>	0.030 <sup>c</sup>	2.56	3.02 <sup>b</sup>	0.027 <sup>c</sup>	2.94

<sup>a</sup> D. J. Hughes, *Nucleonics* **17**, 132 (1959).

<sup>b</sup> Calculated from thermal value and  $d\nu/dE = 1/8 \text{ MeV}^{-1}$ . See R. B. Leachman, *Phys. Rev.* **101**, 1005 (1956).

<sup>c</sup> J. C. Hopkins and B. C. Diven, *Nuclear Sci. and Eng.* **12**, 169 (1962).

<sup>d</sup> R. L. Macklin, G. de Saussure, J. D. Kington, and W. S. Lyon, *Nuclear Sci. and Eng.* **8**, 210 (1960).

<sup>e</sup> R. L. Macklin, G. de Saussure, J. D. Kington, and W. S. Lyon, *Nuclear Sci. and Eng.* **14**, 101 (1962).

in the table serve mainly to reinforce the view held since the earliest days of the Metallurgical Project that breeding in the U-Th cycle ought to be possible at thermal energy and in the U-Pu cycle at high energy. The U-Th cycle is marginal, compared to U-Pu, at high energy; neither cycle looks very good at intermediate energy ( $\sim 30 \text{ keV}$ ).

#### Fast Breeders

Most of the world's work on breeders is centered on U-Pu fast reactors since  $\eta$  is so large under the conditions prevailing in such a reactor. The first feasibility studies of the fast breeder were carried out at the Metallurgical Laboratory by Soodak under Professor Wigner's direction.<sup>49</sup> The technology has proved to be so difficult and slow moving that few innovations not mentioned in this early work have been tried as yet. Four of the world's fast breeders—EBR-1 (1 MW), EBR-II (60 MW), Dounreay (50 MW), Fermi (350 MW)—use liquid-metal cooling and metallic fuel elements as suggested in Soodak's original paper; two others, the Russian BR-5 (5 MW) and the French Rapsodie (10 MW) are fueled with stainless steel-clad  $\text{PuO}_2$ .

<sup>47</sup> J. C. Hopkins and B. C. Diven, *Nuclear Sci. and Eng.* **12**, 169 (1962).

<sup>48</sup> R. L. Macklin, G. de Saussure, J. D. Kington, and W. S. Lyon, *Nuclear Sci. and Eng.* **8**, 210 (1960).

<sup>49</sup> H. Soodak, Chicago Metallurgical Laboratory Report CF-3107 (July 1945) (unpublished).

Although all of the high-powered fast breeders have encountered delays (only Dounreay, BR-5, and EBR-I are operating at power at the time of this writing, May 1962, and the power of Dounreay is only 1/5 of its rated 50 MW), there is no reason to believe that the fast breeders, as reactors, will not operate satisfactorily. But a breeder must not only chain react smoothly, it must also lend itself to easy reprocessing of its partially spent fuel, and this problem remains formidable. Metallic fuel elements such as are used in EBR-II (alloy of uranium plus a mixture of rare earths) can sustain a burnup of about 1% of all atoms, the exact number depending on the temperature at which the reactor operates. Suppose the fuel contains one atom of  $\text{Pu}^{239}$  for every ten atoms of uranium; then before a gram of  $\text{Pu}^{239}$  has been completely burned, it must be recycled ten times. Unless such recycling (including metallurgical refabrication) can be done at, say, 50¢ to \$1.00 per gram, the additional cost, not to say the loss of fissile material, associated with all this handling would make fast breeders uneconomical.

Several ways to improve the situation are being studied, two of which will be mentioned. On the one hand, pyrometallurgical methods such as zone refining are being tried, especially at the EBR-II. Another approach is the use of oxide fuel elements, first tried in the Russian BR-5. Mixtures of  $\text{UO}_2$  and  $\text{PuO}_2$  can withstand burnups of possibly as much as 5%. Thus in an oxide fast breeder one avoids the difficulty of frequent and costly recycle. But one must pay dearly in conversion ratio: First, the presence of the relatively light oxygen degrades the neutron spectrum; second, the fission products, which absorb neutrons, are not removed; and third, the oxide, being less dense than metal, competes with cladding materials less favorably for neutrons than does the metal. For all these reasons the breeding ratio in a fully engineered oxide Pu-U fast breeder is estimated to be about 0.3 less than in a metal-fueled breeder.<sup>50</sup>

#### Slow Breeders

The slow (or thermal) breeders, based on U-Th, though they suffer from the handicap of low  $\eta$ , ought to be relatively freer of the recycle problem that bedevils the fast breeder. In a slow neutron breeder one is at liberty to dilute the fissile material with a low-cross-section ceramic, such as graphite, or a liquid, such as heavy water. In the solid-fueled thermal breeder the concentration of fissile atoms in the fuel

<sup>50</sup> U. S. Atomic Energy Commission Division of Technical Information, *Power Reactor Technology* (September 1961), Vol. 4, p. 84.

is so low that the initial fuel can be burned completely without causing serious radiation damage; moreover, the great dilution of the fissile atoms with innocuous graphite allows the fuel to be burned at very high heat ratings (300 kW/kg). This is the basic rationale for the various graphite, enriched-uranium, gas-cooled reactors such as the German pebble bed, the Dragon (Winfrith Heath), and the High Temperature Gas Reactor (HTGR) (Peach Bottom, Pennsylvania). Reactors of this type were first suggested by Farrington Daniels at the Metallurgical Laboratory in 1944,<sup>51</sup> and fairly serious work was undertaken on them in 1946 at the Clinton Laboratories under Professor Wigner. The systems did not appear attractive at that time, largely because  $U^{235}$  was so expensive then—about \$35/gram. Even if the entire initial load could be burned without reprocessing (as is now expected at both Dragon and HTGR), the burnup charge alone—about 5 mills/kWh—was much too high to be interesting; it was for this reason, fundamentally, that the project was dropped in 1947. The scheme has since been revived because  $U^{235}$  has become cheaper, and because such systems should have an over-all conversion ratio approaching unity. However, the graphite solid-fuel reactors, even using  $U^{233}$ , probably cannot be made into true breeders with breeding ratio  $> 1$ . The main losses are to the graphite and to the fission products, notably  $Xe^{135}$ , which presumably remain in a solid fuel element. The incentive therefore remains for developing a thermal reactor from which fission products (including  $Xe^{135}$ ) are continually removed, which allows easy fuel recycle, and which avoids excessive losses to the moderator.

The aqueous homogeneous reactor (solution of  $UO_2SO_4$  in  $D_2O$ ) satisfies these conditions; and on this account it was given much attention by Wigner and his group at the Metallurgical Laboratory. Several aqueous homogeneous reactor experiments have been built, the most elaborate being the HRE-2, a 5000 kW reactor which operated at Oak Ridge for a total of 12 082 hours between 1957 and 1961. At high temperature and high power it was found that uranium segregated from the HRE-2 solution and deposited on the wall of the inner tank (which was made of zirconium). The deposited uranium overheated and several holes were melted in the wall of the tank.

This difficulty of HRE-2 is traceable to the phase properties of the  $UO_2SO_4$ - $D_2O$  system. Uranyl sulfate becomes *less* soluble as the temperature increases;

<sup>51</sup> Farrington Daniels, Metallurgical Laboratory report N-1668b (October 1944) (unpublished).

above about 320°C uranium-containing solids or heavy liquids separate from solution. Thus one is faced with an unstable situation; if a wall in contact with the fissioning solution inadvertently overheats, uranium tends to deposit, causing further heating and burnout. In principle, by proper hydrodynamic design and careful temperature control one ought to be able to keep all surfaces at a tolerable temperature. If this can in fact be done, the aqueous homogeneous system would be a feasible path to a thermal breeder.

But the problem is difficult, and for this reason other liquid uranium systems, in which the uranium becomes *more* soluble as the temperature rises, are being studied. The most thoroughly examined such system is based on molten fluorides. Mixtures of salts, such as  $Li^7F$ - $NaF$ - $ZrF_4$ - $UF_4$ - $ThF_4$ , melt at about 450°C and remain homogeneous solutions up to 2000°C and beyond. At temperatures of 650°C, which would be the temperature of a power reactor, the salts can be held in molybdenum-nickel alloys or in graphite. Since the salts dissolve thorium as well as uranium fluorides (or plutonium fluoride, for that matter), they ought to be useable in breeder systems. However, to achieve a breeding ratio above one, no high-cross-section structural material can be tolerated inside the reactor. A possible molten salt breeder reactor might therefore consist of a matrix of bare graphite through which the salt, containing both thorium and uranium, would be pumped. The salt would be in direct contact with the graphite, and the feasibility of such a system depends upon how compatible molten fluorides and graphite are under reactor conditions. Without radiation, there seems to be little problem; however, in the presence of radiation, there is evidence that  $UF_4$  in the salt reacts with graphite to form small amounts of  $CF_4$ . It is too early to say how serious this reaction is. In any case plans are well under way to construct a 10 MW Molten Salt Reactor Experiment at Oak Ridge. If this experiment is successful, the way to at least one kind of thermal breeder would seem to be open.

### Research Reactors<sup>52</sup>

Finally, we make a few remarks about research reactors, to whose technology Wigner has contributed so much. Research reactors have become the most popular pieces of very expensive scientific machinery; there are in existence about 260 research reactors, ranging in power from a few watts to 200 megawatts and in price from \$10<sup>5</sup> to \$58 × 10<sup>6</sup>. Of the world's

<sup>52</sup> T. E. Cole and A. M. Weinberg, *Ann. Rev. Nuclear Sci.* (to be published).

research reactors, fully 60% use fuel elements consisting of parallel plates of aluminum containing an alloy of  $U^{235}$ -Al. The prototype of this fuel element was originally developed, largely under the direction of Professor Wigner, for the Materials Testing Reactor.

Modern high-powered research reactors fall into two classes—those moderated with heavy water and those moderated with light water. The light-water reactors are simpler than are the heavy water ones, and are easier to operate at extremely high powers. The Materials Testing Reactor (MTR), which was the first of the light-water-moderated research reactors, ushered in the era of research reactors of the  $10^{14}$  neutrons/cm<sup>2</sup>/sec class. The MTR operates at 40 MW; there are now about a dozen other reactors which are variants of the MTR and which reach slow neutron fluxes up to  $4 \times 10^{14}$ .

Several reactors are now on the drawing board, or under construction, that are expected to reach slow neutron fluxes in excess of  $10^{15}$ . At a slow neutron flux of, say,  $5 \times 10^{15}$ , which is the maximum flux to be achieved in the projected High Flux Isotope Reactor,  $U^{235}$  undergoes fission at the rate of 250 kW/g; about 30% of the  $U^{235}$  is burned per day. A reactor using such highly rated solid fuel elements would be shut down most of the time for changing fuel, assuming even that one could extract the heat when it is produced at so prodigious a rate. Thus to achieve  $5 \times 10^{15}$  flux one must either refuel continuously by using a liquid fuel or a complicated on-line reloading mechanism; or one must separate the region of very high flux from the region occupied by fuel.

The latter solution to the problem of ultra-high flux reactors has been adopted in three of the newest and most advanced research reactors: the Advanced Testing Reactor (250 MW) to be built in Idaho, the Soviet S. M. Reactor (50 MW), and the High Flux Isotope Reactor (100 MW) to be built in Oak Ridge. In these reactors, the fuel is disposed as an annulus around a weakly absorbing island, called a "flux-trap." Since the lifetime of neutrons in the island may exceed the lifetime in the fuel-bearing annulus

by a factor of ten or so, the slow-neutron flux in the trap can exceed that in the annulus by a corresponding factor. In this way one achieves a local slow-neutron flux of  $5 \times 10^{15}$  while the flux in the fuel remains well below  $10^{15}$ .

Whether research reactors in the  $2$  to  $5 \times 10^{16}$  neutrons/cm<sup>2</sup>/sec class are feasible is still a moot point. Even with an extreme flux trap, parts of the fuel would be exposed to fluxes of several times  $10^{15}$ . The fuel would be burned so rapidly as to make a solid-fueled reactor a doubtful proposition. On the other hand, quick turnover of fuel is no problem if the fuel is liquid. Research reactors in the  $10^{16}$  flux class might well be by-products of successful development of very high power density liquid-fueled reactors.

#### SUMMARY

In this short review we have tried to give the reader a feeling for the pervasiveness of Eugene Wigner's contributions both to the underlying theory and to the engineering development of chain reactors. The theoretical topics which we chose to discuss—the spectrum of the Boltzmann equation, neutron thermalization, thermal utilization, resonance absorption—exhaust neither the recent significant theoretical developments nor Wigner's contributions thereto. This is even more the case with respect to reactor technology; Wigner has contributed to many parts of the technology in addition to the ones we have described. Since chain reactor technology is so massive and slow moving, it will probably take a generation to decide on the correctness of the fundamental strategy espoused by Professor Wigner—that the development of the power breeder is the central technological problem of nuclear energy. Nevertheless, those of us who have worked with Professor Wigner ever since the beginning of the atomic age and who have always been amazed at his uncanny technical prescience look forward to the happy occasion of, say, Professor Wigner's eightieth birthday when the full technological validity of his views on breeders will undoubtedly have been amply demonstrated.