The behavior of the neutron energy spectrum in a bare homogeneous moderator after the introduction of a burst of neutrons has been studied theoretically. The problem is reduced to an eigenvalue problem for a self-adjoint integral operator governing the neutron thermalization.

In an infinite system, it has been shown that the Maxwellian component of the spectrum does not couple with higher modes which are orthogonal to the former and contribute nothing to the actual neutron density when integrated. The asymptotic neutron spectrum and the diffusion cooling are formulated most conveniently in the present scheme.

The formalism is a general one and may be applied to any scattering model with due labor of computations. Numerical calculations has been done for heavy gaseous moderators.

**INTRODUCTION**

The behavior of the neutron energy spectrum after the introduction of a burst of neutrons in a moderator is of considerable interest in the analysis and interpretation of pulsed neutron experiments. Several authors have derived the decay constant and the spectrum approached after the transient components have decayed, but few were concerned with the transient itself. Concerning the transient, attention has been paid solely on the ground modes of decay. Associated with each of the spatial higher harmonics, however, there exist energetically higher modes of decay as well as the ground mode. And even if a burst of neutron with the spatial distribution corresponding with a single spatial harmonics were injected initially, the neutron spectrum would not stand invariably but would shift towards the softer side. In fact, the time-dependent neutron spectrum in a finite system should be considered consisting of characteristic spectra, each having an independent mode of decay.

The concept of characteristic spectra has been introduced and studied by Kazarnovsky, Katsuragi, and Purohit, but has not been developed satisfactorily by them. The author proposes here a general formalism in which the characteristic spectra are determined as eigenfunctions of a self-adjoint integral operator governing the thermalization and operating in the function space composed of modified density spectra. The eigenvalue problem can be solved approximately through the Rayleigh-Ritz variational method by expanding the modified density spectrum into a suitable orthonormal series.

Most important case relates to an infinite system. Here the ground mode, i.e., the Maxwellian spectrum, persists without being coupled with energetically higher modes which contribute nothing to the actual neutron density when integrated over the whole energy range. Although the energetically higher modes are expected to reveal the thermalizing property of a moderating material, their detection is not easy, contrary to Purohit's suggestion, because usual 1/ν-detectors are useless for the detection.

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**研究論文**

238. Characteristic Spectra in Neutron Thermalization

熱中性子化の特性スペクトル

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However an essential information on neutron thermalization will surely be obtained by the analysis of the problem in the infinite system. The problem in the finite system can be dealt with by the perturbation technique utilizing the results for the infinite system.

Thus, in particular, the asymptotic spectrum and the diffusion cooling coefficient find their most adequate expressions in the present scheme. The inverse proportionality of the diffusion cooling coefficient to the first higher decay constant, as reported by Purohit, is shown to be merely the first approximation. The role played by the second moment of the energy transfer kernel in the neutron thermalization may be understood properly in the present scheme.

I. Basic Equations

In the diffusion approximation, the neutron flux $\phi(r, E, t)$ in a bare homogeneous moderator obeys Eq. (1):

$$\frac{1}{v} \frac{\partial \phi(r, E, t)}{\partial t} = D(E) \nabla^2 \phi(r, E, t) - \sum_\alpha N_\alpha(E, t) \phi(r, E, t) + \int_0^E \sum_\alpha (E' \rightarrow E) \phi(r, E', t) dE' + S(r, E, t).$$

Attention is devoted here to the case where the source is a burst of neutrons, i.e., $S(r, E, t) = S(r, E) \delta(t)$. Take a set of spatial harmonics, $X_m(r)$, $m = 0, 1, 2, \ldots$, vanishing on the extrapolated boundary of the system and obeying Eq. (2):

$$\nabla^2 X_m(r) + B_m^2 X_m(r) = 0.$$ (2)

Then the neutron flux $\phi$ is expanded into a series of the form:

$$\phi(r, E, t) = \sum_m v_m N_m(E, t) X_m(r).$$ (3)

Insertion of the expression (3) into Eq. (1) yields an equation obeyed by the neutron energy density spectrum $N_m(E, t)$ accompanied by the $m$-th spatial harmonics $X_m(r)$:

$$\frac{1}{v} \frac{\partial N_m(E, t)}{\partial t} = D(E) B_m^2 v_m E^3 N_m(E, t) + \sum_\alpha v_\alpha N_\alpha(E, t) + v_0 \sum_\alpha (E' \rightarrow E) E^4 N_m(E, E', t) + S(r, E, t).$$ (4)

where $E$ is the energy in $kT$ unit, $v_0$ the neutron velocity corresponding to $E = kT$, and $\sum_\alpha$ the macroscopic absorption cross section for $v = v_0$. The absorption cross section is assumed to be inversely proportional to the neutron velocity throughout the paper.

Assuming a separable solution of the form

$$N_m(E, t) = N_m(E) e^{-\lambda_m t},$$ (5)

the mode of spectrum $N_m(E)$ and its associated decay constant $\lambda_m$ should be determined by an integral equation:

$$\lambda_m - \sum_\alpha v_\alpha N_\alpha(E) = D(E) B_m^2 v_0 E^3 N_m(E) + v_0 \int_0^E \sum_\alpha (E' \rightarrow E) E^4 N_m(E') dE'. $$ (6)

The problem has thus been reduced to an eigenvalue problem for the operator:

$$D(E) B_m^2 v_0 E^3 \cdot + v_0 \sum_\alpha (E' \rightarrow E) E^4 \cdot - v_0 \int_0^E dE' \sum_\alpha (E' \rightarrow E) E^4 N_m(E') dE'. $$ (7)

The kernel in this operator is polar, and hence the operator can be made symmetric by introducing the modified density spectrum:

$$n(E) = \frac{E^4}{\sqrt{M(E)}} N(E),$$ (8)

where $M(E)$ denotes the Maxwellian flux distribution, $E e^{-E}$. The modified density spectrum $n(E)$ now obeys an integral equation:

$$v n(E) = H n(E),$$ (9)

where $\nu$ is the new eigenvalue connected with $\lambda$ by

$$\nu = \lambda - \sum_\alpha v_\alpha v_0,$$ (10)

and $H$ stands for the operator:

$$H = D(E) B_m^2 E^3 \cdot + v_0 \sum_\alpha (E') E^4 \cdot - v_0 \int_0^E dE' \sum_\alpha (E' \rightarrow E) E^4 \sqrt{M(E')}.$$ (11)

which is symmetric in virtue of the condition of detailed balance:

$$M(E') \sum_\alpha (E' \rightarrow E) = M(E) \sum_\alpha (E \rightarrow E').$$ (12)

In Eqs. (8)~(11), the index $m$ has been dropped for simplicity, and hence $B^2$ in the expression (11) for the operator $H$ should be read as $B_m^2$ hereafter.

Now that the operator $H$ is self-adjoint, its eigenvalues are equivalent to the stationary values of the functional $J$:

$$J(\nu) = \frac{\langle \phi, H \phi \rangle}{\langle \phi, \phi \rangle},$$ (13)
where \( n_T(E) \) is a trial function, and the parenthesis \( (,\,) \) denotes an inner product of functions:

\[
(x, y) = \int_0^\infty x(E) \cdot y(E) \, dE.
\]

To solve the variational problem for the functional \( J \), the Rayleigh-Ritz method is most straightforward. Here the trial function \( n_T(E) \) is expanded in terms of suitable orthonormal bases \( \omega_j(E) \), thus

\[
n_T(E) = \sum_j c_j \omega_j(E).
\]

This leads to a secular equation for the eigenvalue \( \nu \):

\[
det | \lambda_{jk} - \nu \delta_{jk} | = 0,
\]

where \( \lambda_{jk} = (\omega_j, H \omega_k) = \alpha_{kj} \).

When the expansion is truncated at the \( n \)-th term, the \( n \) roots, \( \nu_0 < \nu_1 < \nu_2 \ldots < \nu_{n-1} \), of the secular equation give approximate values, upper bounds, of the true eigenvalues. The secular Eq. (15) may readily be solved numerically according to standard computer codes. The component \( c_j \) of the eigenvector, accompanying with each eigenvalue, gives an approximation to the true amplitude of \( \omega_j(E) \) in the modified spectrum \( n(E) \). Naturally the eigenvector is assumed to be normalized, so that

\[
\sum_j c_j^2 = 1,
\]

or \( (n_T, n_T) = 1 \).

Hereafter use is made of the notations \( \nu_{mn} \) and \( n_{mn}(E) \) for the \( n \)-th eigenvalue and its associated characteristic spectra of Eq. (9) having the operator \( H \) with \( B^2 \) replaced by \( B_{2m}^2 \). It is clear that the \( m \)-th mode of the neutron spectrum is given by a superposition of the form:

\[
N_n(E, t) = \sum_n A_{mn} N_{mn}(E) e^{-\lambda_{mn} t},
\]

where

\[
N_{mn}(E) = \sqrt{\frac{M(E)}{E^3}} n_{mn}(E)
\]

and \( \lambda_{mn} = \nu_{mn} + \sum_0 \nu_0 \).

The expressions (3), (17) and (18) solve formally the time-dependent problem. The amplitudes, \( A_{mn} \), of each characteristic mode of spectrum should be determined by the initial condition rather than by properties of the material.

### II. Characteristic Modes of Spectra

1. Orthogonality

Any two energy eigenfunctions of Eq. (9) belonging to different eigenvalues are orthogonal to each other:

\[
\int_0^\infty n_n(E) n_m(E) \, dE = \delta_{nm},
\]

where \( N^*_n(E) \) is the adjoint of the characteristic spectrum \( N_n(E) \):

\[
N^*_n(E) = \frac{E^3}{M(E)} N_n(E) = -\frac{E^3}{\sqrt{M(E)}} n_n(E).
\]

The orthogonality holds for any fixed value \( B_{2m}^2 \).

2. The Maxwellian spectrum in the infinite system

By physical intuition, one is convinced of the basic situation of the Maxwellian spectrum in the infinite system. In fact, one sees directly that Eq. (1) has a separable solution \( \phi(E, t) = M(E) \exp(-\sum_0 \nu_0 t) \), or equivalently that Eq. (4) admits a separable solution (5) with \( N_n(E) \) replaced by \( E^{-1/2} M(E) \), so long as the infinite system is composed of \( 1/\nu \)-absorbers.

It is worth noticing that this is the case irrespectively of whether the moderating material is composed of free atoms or of chemically bound ones.

Now the Maxwellian mode is written as \( E^{-1/4} \sqrt{M(E)} = E^{1/4} e^{-\xi/2} \) in the function space of modified density spectra. This suggests the convenience of adopting, as the orthonormal bases in Eq. (14), the associated Laguerre polynomials of order \( 1/2 \), \( L_{j}^{(1/2)}(E) \) with the weight \( E^{1/4} e^{-\xi/2} \):

\[
\omega_j(E) = \sqrt{\frac{j+1}{\Gamma(j+1/2)}} E^{1/4} e^{-\xi/2} L_{j}^{(1/2)}(E)
\]

where the numerical coefficient is the normalization factor. It may be recognized that the spectrum represented by \( \omega_j(E) \) grows hard with increasing \( j \).

By choosing a set of bases as given in Eq. (21), one can write the secular equation

\[
\sum_j c_j^2 = 1, \quad (n_T, n_T) = 1.
\]
(15), for the infinite system, in the following form:

\[
\begin{pmatrix}
-\nu & 0 & 0 & 0 & \cdots \\
0 & a_{11} - \nu & a_{12} & a_{13} & \cdots \\
0 & a_{21} & a_{22} - \nu & a_{23} & \cdots \\
0 & a_{31} & a_{32} & a_{33} - \nu & \cdots \\
\vdots & \vdots & \vdots & \vdots & \ddots
\end{pmatrix} = 0,
\]

(22)

which shows that the ground mode, the Maxwellian spectrum, does not couple with higher modes, as it should. Eq. (22) is consistent with those facts that the fundamental decay constant is given by

\[
\lambda_0 = \sum_{n} \epsilon_n v_0,
\]

(23)

and that its associated characteristic spectrum is the Maxwellian:

\[
N_0(E) = \frac{\sqrt{\lambda_0 / \pi} }{E} n_0(E) = \frac{2}{\sqrt{\pi}} \frac{M(E)}{E},
\]

and

\[
N_0^+(E) = \frac{2}{\sqrt{\pi}},
\]

(24)

where \(\sqrt{2/\sqrt{\pi}}\) is the normalization factor.

3. The Higher Modes in the Infinite System

From Eq. (22), it is seen that any higher modes \(n_n, n > 1\), of the modified spectrum contains no Maxwellian mode, \(n_0\), and are written in the form:

\[
n_n(E) = \sum_{k=1}^{\infty} c_n k \left( \frac{k!}{\Gamma\left(k + \frac{3}{2}\right)} \right) \frac{\sqrt{M(E)}}{E} L_k^{(4)}(E),
\]

(25)

This is rewritten in the form of the density spectrum as

\[
N_n(E) = \sum_{k=1}^{\infty} c_n k \left( \frac{k!}{\Gamma\left(k + \frac{3}{2}\right)} \right) \frac{\sqrt{M(E)}}{E} L_k^{(4)}(E),
\]

(26)

showing that the "higher" mode is composed of "energetically harder" spectra than the Maxwellian density spectrum \(N_0(E) = E^{-1/2} M(E)\).

It may be remarked here that higher modes of the density spectrum in the infinite system contribute nothing to the actual neutron density, since it is integrated to be zero:

\[
\int_0^\infty N_n(E) dE = \frac{\sqrt{\pi}}{2} \int_0^\infty N_n^+(E) N_n(E) dE = 0,
\]

(27)

because of Eqs. (19) and (26). This means, in turn, that the energetically higher modes of decay in the infinite system cannot be detected by using \(1/\nu\)-detectors. Thus, Purohit's suggestion to utilize the first energetically higher mode of decay in the study of the thermalizing property of the material is believed powerless unless a new contrivance of other detector than the conventional \(1/\nu\)-type is made.

However, this does not imply that the higher modes of decay are in imaginary existence. It does exist always accompanying the Maxwellian mode which is approached asymptotically, and the decay of these energetically higher modes of spectra is nothing but the thermalization process of neutrons.

III. HEAVY GASEOUS MODERATORS

As a simplest example, heavy gaseous moderators are studied in this chapter. For convenience, the elements of the determinant in Eq. (15) are divided into two parts:

\[
a_{nj} = \langle \omega_j, H \omega_n \rangle = \langle \omega_j, D(E) v_0 B^2 E^4 \rangle \langle \omega_n \rangle
\]

\[
+ \langle \omega_j, H \omega_n \rangle = W_{nj} + V_{nj}.
\]

(28)

The diffusion constant \(D\) may be regarded as constant in the heavy gas model, and \(W_{nj}\) is evaluated to be

\[
W_{nj} = D v_0 B^2 \frac{k!}{\Gamma\left(j + \frac{3}{2}\right) \Gamma\left(k + \frac{3}{2}\right)} \frac{\sqrt{M(E)}}{E} \left\{ \sum_{r=0}^{n} \frac{\Gamma\left(j - \tau - \frac{1}{2}\right)}{\Gamma\left(k - \tau - \frac{1}{2}\right)} \Gamma\left(\tau \frac{1}{2}\right) \right\},
\]

(29)

where the summation over \(r\) extends from 0 to \(j\) or \(k\), whichever is smaller.

The calculation of \(V_{nj}\) is based on the fact that

\[
H \omega_n = v_0 \frac{k!}{\Gamma\left(k + \frac{3}{2}\right) \sqrt{M(E)}} \times R\{M(E) L_k^{(4)}(E)\},
\]

(30)

where \(R\) is the operator defined by

\[
R = \Sigma_n (E) \cdot - \int_0^\infty dE' \Sigma_n (E' \rightarrow E'),
\]

(31)

It is well known that when the higher-order
terms in inverse mass are neglected, $R$ is approximated by the differential operator:

$$R = -\xi \sum_{\alpha} \left( E \frac{d^2}{dE^2} + E \frac{d}{dE} + 1 \right),$$

which admits as its eigenfunctions the associated Laguerre polynomials of order 1 multiplied by $M(E)$:

$$RM(E) L_n^{(1)}(E) = n \xi \sum_{\alpha} M(E) L_n^{(1)}(E),$$

where $\xi$ is the average logarithmic energy decrement per collision and $\sum_{\alpha}$ is the scattering cross section for free atom. Now, a reflection upon the relation:

$$L_n^{(4)}(E) = \sum_{r=0}^{\infty} \frac{\Gamma(r - \frac{1}{2})}{r! \Gamma(-\frac{1}{2})} L_r(E),$$

make the operation $R \cdot$ in Eq. (30) an easy algebra:

$$H_{\alpha \beta} = \xi \sum_{\alpha} v_{\alpha} \left\{ \frac{k!}{\Gamma(k + \frac{3}{2})} \right\} \cdot M(E)$$

$$\sum_{r=0}^{\infty} \left( k - r \right) \frac{\Gamma(r - \frac{1}{2})}{r! \Gamma(-\frac{1}{2})} L_r^{(4)}(E).$$

Thus, one obtains

$$V_{j \alpha} = \xi \sum_{\alpha} v_{\alpha} \left\{ \frac{j!}{\Gamma(j + \frac{3}{2})} \right\} \frac{k!}{\Gamma(k + \frac{3}{2})} \Gamma(k - r - \frac{1}{2}) \Gamma(-\frac{1}{2})$$

$$\times \left( \sum_{r=1}^{\infty} \frac{\Gamma(j - r - \frac{1}{2})}{(j - r)! (k - r)! \Gamma(-\frac{1}{2})} \right) \times r (r + 1)$$

where the summation over $r$ extends from 1 to $j$ or $k$, whichever is smaller.

It is to be noted here that two parts of the element of the determinant, $W_{j \alpha}$ and $V_{j \alpha}$, are connected by a relation:

$$V_{j \alpha} = \frac{2 \xi \sum_{\alpha} W_{j \alpha}}{DB^2} \left[ W_{j-1, k-1} + W_{j-2, k-2} + \cdots \right.$$

$$+ W_{0, k-1} + k \geq j,$$

so that, in practice, only $W_{j \alpha}$'s are required to be evaluated.

The secular equation (15) for several values of $B^2$ have been solved numerically by retaining the first ten terms in the expansion (14), with $w_j(E)$ given by Eq. (21). Lower eigenvalues and their associated eigenfunctions in the infinite system are shown in Table 1 and in Fig. 1. Fig. 2 shows the variation in eigenvalues with increasing spatial buckling $B^2$.

The range of the buckling $B^2$ extends from 0 to $DB^2/\xi \sum_{\alpha} = 1$, which corresponds roughly to the smallest allowable size for the diffusion approximation to be valid. The dotted line represents the eigenvalues derived by neglecting the deviation of the asymptotic spectrum from the Maxwellian. Therefore, the difference between the curve of $\lambda$ vs. $B^2$ and the dotted line represents a theoretical

<table>
<thead>
<tr>
<th>Order</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eigenvalue divided by $\xi \sum_{\alpha} v_{\alpha}$</td>
<td>0</td>
<td>1.189</td>
<td>2.756</td>
<td>4.994</td>
<td>8.016</td>
</tr>
</tbody>
</table>

Table 1: Eigenvalues in the infinite system of heavy gaseous moderator
(Calculated by retaining the first ten terms in the expansion (14), with $w_j(E)$ given by Eq. (21))

![Fig. 1 Characteristic spectra in the infinite system of heavy gaseous moderator](image1.png)

![Fig. 2 Variation in eigenvalues with increasing spatial buckling](image2.png)
prediction of the diffusion cooling effect in heavy gaseous moderators, which allows one to compare directly with the experimental results. Softening of asymptotic spectra caused by the diffusion cooling is shown in Fig 3.

Fig. 3 Softening of asymptotic spectra caused by the diffusion cooling

From the results obtained, eigenvalues are found to be distributed at increasing intervals with increasing order. Eigenvalues for the heavy gaseous moderator are proportional to $\xi\Sigma\rho_0$, so that the increment of the mass of the moderating atom makes the eigenvalues smaller, whereas the eigenfunctions remain unchanged provided $DB^3/\xi\Sigma_\alpha$ is kept constant.

IV. FORMULATIONS IN CHARACTERISTIC MODES OF SPECTRA

1. A general description of the thermalization process

The analysis in Chap. I shows that the thermalization of neutrons in a bare homogeneous moderator proceeds according to Eq. (17) or (38):

$$N(r, E, t) = \sum_n \sum_\alpha A_{n\alpha} e^{-\lambda_{n\alpha}t} N_{n\alpha}(E) X_n(r).$$  (38)

The amplitudes, $A_{n\alpha}$, of each mode are determined by the initial condition:

$$A_{n\alpha} = \int_0^\infty N_{n\alpha}^*(E) dE \int \frac{X_n(r)N(r, E, 0)dr}{\text{Volume}}.$$  (39)

Since it is not practical to expand the high energy spectrum of the source directly in terms of the characteristic modes of thermalization, a good recommendation will be to replace $N(r, E, 0)$ in Eq. (39) by an appropriate epithermal spectrum, which is derived through the analysis of the slowingdown process starting from the original source spectrum. This technique is useful in the next section also.

When the source neutrons are not pulsed but are given by a continuous function $S(r, E, t)$, the neutron spectrum in the system is described by

$$N(r, E, t) = \sum_\alpha \sum_n A_{n\alpha} e^{-\lambda_{n\alpha}t} \int S_{n\alpha}(t') e^{-\lambda_{n\alpha}(t-t')} dt'.$$  (40)

where

$$S_{n\alpha}(t') = \int_0^\infty N_{n\alpha}^*(E) dE \int X_n(r) S(r, E, t') dr.$$  (41)

2. Equilibrium spectrum in the infinite system with a steady source

In case of the infinite system with a steady and uniform source, Eq. (40) reduces to

$$N(E) = \sum_\alpha \sum_n S_{n\alpha} N_{n\alpha}(E) = \sum_\alpha \frac{S_{n\alpha}}{\nu_n + \sum_{\alpha=1}^\infty \nu_{n\alpha}} N_{n\alpha}(E),$$  (42)

where

$$S_{n\alpha} = \int_0^\infty N_{n\alpha}^*(E) S(E) dE.$$  (43)

Thus, the equilibrium spectrum is written as a superposition of characteristic spectra. One sees that the equilibrium spectrum (42) presents some qualitative features: i) Attainment of the Maxwellian spectrum in case of no absorption, ii) Increment of the proportional weight of the higher modes in case of strong absorption, iii) Increment of the deviation from the Maxwellian mode with increasing mass of the moderating atom. The feature iii) comes from the fact that the increase in the mass makes $\xi$, and hence $\nu_n$'s but $\nu_0$, smaller.

Furthermore, Eq. (42) and the summational nullity of the higher modes discussed in Chap. II lead to the conclusion that the total absorption in the infinite system is, in fact, ascribable solely to the Maxwellian mode of the spectrum so long as the absorption obey the $1/v$-law. Now the effective absorption cross section $\Sigma_{\text{eff}}$ is defined by

$$\Sigma_{\text{eff}} = \frac{\int \Sigma_n(E) \phi(E) dE}{\int \phi(E) dE}.$$  (44)
In case of \(1/v\)-absorption, the numerator is reduced to \(\sum a_0v_0N\), where \(N\) is the total neutron density and is invariant independently of amplitudes of higher modes, whereas the denominator, the total neutron flux, increases as amplitudes of higher modes increase.

Thus, the role played by higher modes is to raise the value of the total flux and to diminish the effective cross section, under the condition of constant neutron density and constant absorption rate. This is nothing but the cause of the hardening of the spectrum.

3. Diffusion cooling

The asymptotic spectrum in the finite system with a pulsed source is also worth studying. In fact, the diffusion cooling effect can be examined by drawing the \(\lambda\) vs. \(B^2\) curve similar to Fig. 2 using a suitable scattering model for the material.

The expression for the diffusion cooling coefficient in terms of properties of the moderating material is obtained by the perturbation technique. The self-adjoint operator \(H\) in Eq. (11) is here divided into two parts:

\[
H = H_0 + B^2 V, \quad (45)
\]

where \(B^2 V\) is the perturbation and

\[
V = D(E)\nu_0 E^4. \quad (46)
\]

Assume that the eigenvalue problem for the operator \(H_0\) has already been solved:

\[
H_0\nu_n = \nu_n\nu_n, \quad n = 0, 1, 2, \ldots. \quad (47)
\]

Then the lowest eigenvalue \(\nu\) and its associated eigenfunction \(\nu\) of the operator \(H\) are obtained through the usual procedure.

\[
\nu = \nu_0 + B^2 (V\nu_0, \nu_0 - B\sum_{n=0}^{\infty} (V\nu_n, \nu_n)^2 + O(B^4). \quad (48)
\]

\[
\nu = \nu_0 - B^2 \sum_{n=1}^{\infty} (V\nu_n, \nu_n) - \nu_0 - \nu_n + O(B^4). \quad (49)
\]

Recalling that the ground mode \(\nu_0\) in the infinite system is the Maxwellian and that \(\nu_0 = 0\), the fundamental decay constant and its associated asymptotic spectrum in the large finite system are known to be

\[
\lambda = \sum a_0v_0 + B^2 \frac{2}{\sqrt{\pi}} \nu_0 \int_0^{\infty} D(E)M(E) dE dE\nonumber
\]

\[
- B^2 \frac{2}{\sqrt{\pi}} \nu_0 \sum_{n=1}^{\infty} \left( \frac{\int_0^{\infty} D(E)E^4\sqrt{M(E)}n_n(E) dE}{\nu_n} \right)^2 + O(B^4), \quad (50)
\]

\[
n(E) = \frac{\int_0^{\infty} D(E)E^4\sqrt{M(E)}n_n(E) dE}{\nu_n} \quad (51)
\]

\[
\frac{2}{\sqrt{\pi}} \int_0^{\infty} D(E)E^4\sqrt{M(E)}n_n(E) dE \quad (52)
\]

where \(\frac{2}{\sqrt{\pi}}\) is the normalization factor.

The so-called diffusion cooling coefficient is given by the coefficient of \(B^4\) in the above expression for \(\lambda\). When all the terms but the first are neglected in the summation, it reduces to

\[
\frac{2}{\sqrt{\pi}} \nu_0 \left\{ \int_0^{\infty} \frac{1}{\nu_0} \int_0^{\infty} D(E)M(E)L_1(4)(E) dE \right\}^2, \quad (53)
\]

which coincides with Purohit's expression

\[
C = \frac{2}{3} \frac{\nu_0^2}{\pi \nu_1}, \quad (53)
\]

when \(D(E)\) is assumed to be constant.

4. The effect of chemical binding

Compared with the free atomic state, the chemically bound state of moderating materials will relax the thermalization of neutrons. Hence, in the infinite system, the effect of chemical binding is expected to diminish decay constants associated with higher mode of spectra, whereas, as noticed in Chap. II-2, the ground mode is always the Maxwellian and its associated decay constant, \(\lambda_0\), equals \(\sum a_0v_0\) irrespectively of whether the moderator consists of atomic gas or of molecular liquid.

It should be pointed out here that the decay constant of the first higher mode minus \(\sum a_0v_0\), i.e., \(\nu_1\), is roughly proportional to the second moment of the energy transfer kernel. Assuming that the element \(a_{12}, a_{13}, \ldots\) in Eq. (22) are negligibly small, it is seen that the first higher eigenvalue \(\nu_1\) approximates to \(a_{11}\):

\[
\nu_1 = a_{11} = (\nu_1, H\nu_{10})\nonumber
\]

\[
= \frac{\nu_0}{\Gamma\left(\frac{5}{2}\right)} \left\{ \int_0^{\infty} \frac{\nu_0^2 M(E) L_1(4) E L_1(4) E^2 dE}{E^2 E^2} \right\}\nonumber
\]

\[
= \frac{\nu_0}{\Gamma\left(\frac{5}{2}\right)} \left\{ \int_0^{\infty} \nu_0^2 M(E) L_1(4) E L_1(4) E^2 dE\right\} dE\nonumber
\]

\[
\frac{\nu_0}{\Gamma\left(\frac{5}{2}\right)} \left\{ \int_0^{\infty} \nu_0^2 M(E) L_1(4) E L_1(4) E^2 dE\right\} dE, \quad (54)
\]

In virtue of the condition of detailed balance, the wave bracket in the right-hand side of Eq. (54) is shown to be half of the second moment, \(M_2\), of the energy transfer kernel:

\[
M_2 = \frac{1}{\Gamma\left(\frac{5}{2}\right)} \int_0^{\infty} \int_0^{\infty} \nu_0^2 M(E) L_1(4) E L_1(4) E^2 dE dE, \quad (55)
\]

and one obtains

\[
\nu_1 = a_{11} = \frac{2\nu_0 M_2}{3\sqrt{\pi}}. \quad (56)
\]
The approximation to $v_1$ may be improved by taking into account the elements $a_{12}$, $a_{13}$, $a_{22}$, which introduce the third and higher moments of the energy transfer kernel. It is evident that the second moment of the energy transfer kernel, and hence the decay constant of the first higher mode, are smaller for chemically bound atoms than for free atoms. As for the decay constants of the other higher modes, things will go analogously to the first higher mode, and hence they are expected to be diminished also by the effect of chemical binding.

V. DISCUSSIONS AND CONCLUSION

The present formalism is based on a mathematically clear foundation and is believed to give a most convenient scheme among the similar treatments. Important conclusions contain the independence of the Maxwellian mode and the summational nullity of the higher modes in the infinite system composed of $1/v$-absorbers. Higher modes of decay are expected to reveal the thermalizing property of the material more clearly than the usual equilibrium spectrum, but a new contrivance of other experimental technique than the $1/v$-detector is necessary to detect them.

That the eigenvalues are spaced at increasing intervals suggests the importance of the first higher mode of the spectrum, the decay constant of which is the thermalization time constant as pointed out by Purohit, and may be regarded as an indicator of the thermalizing property of the material. On measuring the thermalization time constant for a finite system, care should be employed in distinguishing between two sorts of higher modes, one corresponding to higher spatial harmonics and the other belonging to higher characteristic spectra. In fact, the curves of $\lambda_0$ and $\lambda_1$ in Fig. 2 suggest that the decay constant $\lambda_{01}$ and $\lambda_{11}$ are of the comparable order. (Of $\lambda_m$, the first index $m$ means the spatial mode and the second one $n$ the energetic mode.) For example, the first higher spatial buckling of a long cylinder is 5.36 times as large as the fundamental one. Assuming the value of $DB^2/\Sigma$ $\Sigma_v$ to be, say, 0.2, $\lambda_{10}$ becomes 1.03 $\xi \Sigma_v \Sigma_0$, and $\lambda_{01}$ is 1.41 $\xi \Sigma_v \Sigma_0$, whereas the fundamental decay constant $\lambda_{00}$ has the value of 0.22 $\xi \Sigma_v \Sigma_0$.

Although $\lambda_{01}$ amounts to a higher value than $\lambda_{10}$ in this example, the thermalization time constant, i.e. $\lambda_{01}$, for the finite system is expected to diminish when the chemical binding of the moderating material is taken into consideration. This expectation comes from the fact that $v_1$ is approximately proportional to the second moment of the energy transfer kernel, as mentioned in Chap. I-4.

Eq. (42) has in it another possibility of finding the thermalizing property of the medium. By letting the neutrons having a definite near-thermal spectrum be injected continuously into the medium, and by analyzing the equilibrium spectra induced by various source spectra it might be possible to evaluate the lower eigenvalues. Thereby, the source spectrum should not be so hard, otherwise the equilibrium spectrum resulting from it would not be different widely from the standard one having the $1/E$-tail.

The present formulation must be a standard one in the theory of neutron thermalization, since the formalism is general and may be applied to any scattering model. However, the computational labor will be tedious for molecular moderators, since it involves evaluation of the higher moments of the energy transfer to chemically bound atoms. It is believed that a next step in the theory should be taken towards the investigation of the energy transfer to chemically bound moderators. When this problem can be formulated conveniently by introducing some physical parameters, the theory of neutron thermalization will make a further progress.

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