Letter to the Editor

A New Method for Measuring Thermalization Parameters

The pulsed neutron source technique has been extensively used in the past few years for measuring diffusion parameters of thermal neutrons in moderators (*1, 2*). The nonlinearity in the curve of the decay constant as a function of geometrical buckling is partly due to transport correction to the diffusion approximation (*3, 4*) and partly due to the "diffusion cooling" effect. The latter is related to the energy transfer mechanism between thermal neutrons and moderator. There are some calculations of "diffusion cooling" coefficients based on the theory of inelastic scattering of thermal neutrons, but their meaning is not so clear in view of the large discrepancies in the "diffusion cooling" coefficient measured by several authors.

We propose here another approach to the neutron thermalization problem, namely, analyzing several independent measurements in terms of phenomenological parameters. The present treatment is restricted to homogeneous moderating systems in which "non $1/v$" absorbers can be dispersed, but it can be utilized as a starting point to the more elaborate problem of space dependent thermalization.

The diffusion equation for the flux in a moderator after a short pulse of fast neutrons, assuming time dependence $e^{-\lambda t}$ and taking into account only the lowest mode with geometrical buckling B^2, is

$$\left[\Sigma_a(E) - \frac{\lambda}{v} + B^2 D(E)\right]\phi(E) = \int_0^\infty dE' \cdot[\Sigma_s(E' \to E)\phi(E') - \Sigma_s(E \to E')\phi(E)] \tag{1}$$

where $\phi(E)$ is the flux per unit energy, $\Sigma_a(E)$ is the macroscopic absorption cross section, $D(E)$ is the energy dependent diffusion coefficient, and $\Sigma_s(E \to E')$ is the macroscopic energy transfer cross section for scattering from energy E into a unit energy interval at E'. We assume that the energy spectrum does not deviate strongly from a Maxwellian and we may write it as a Maxwellian times a correction. It is convenient to represent this correction by the set of Laguerre polynomials of order unity and degree n $L_n^{(1)}(E/T)$ where T is the moderator temperature in energy units (*5–7*). The normalization is

$$\int_0^\infty \frac{E}{T^2} e^{-E/T} L_k^{(1)}\left(\frac{E}{T}\right) L_q^{(1)}\left(\frac{E}{T}\right) dE = \delta_{Kq}$$

We write the approximation

$$\phi(E) = M_T(E) \sum_{n=0}^{L} C_n^{(0)} L_n^{(1)}\left(\frac{E}{T}\right) \tag{2}$$

where $M_T(E) = (E/T^2)e^{-E/T}$. The coefficients $C_n^{(0)}$ are determined by the system of homogeneous linear equations

$$\sum_{q=0}^{L} d_{Kq}\, C_q^{(0)} = 0 \qquad K = 0, \cdots L \tag{3}$$

where

$$d_{Kq} = (\Sigma_a)_{Kq} - \frac{\lambda}{v_0}\left(\frac{\sqrt{T}}{\sqrt{E}}\right)_{Kq} + B^2 D_{Kq} + \gamma_{Kq} \tag{4}$$

$$v_0 = \sqrt{\frac{2\,K_B\,T}{M_n}}$$

$$\gamma_{Kq} = \int_0^\infty \int_0^\infty dE\, dE' M_T(E)\Sigma_s(E \to E') \cdot\left[L_K^{(1)}\left(\frac{E}{T}\right) - L_K^{(1)}\left(\frac{E'}{T}\right)\right] L_q^{(1)}\left(\frac{E}{T}\right) \tag{5}$$

and the other terms are defined as follows:

$$f_{Kq} = \int_0^\infty L_K^{(1)}\left(\frac{E}{T}\right) M_T(E) f(E) L_q^{(1)}\left(\frac{E}{T}\right) dE \tag{6}$$

The notation γ_{Kq} is that of Kazarnovsky *et al.* (*5*) and Singwi (*6*). $\gamma_{11} = M_2/4$ where M_2 is the "thermalization power" (*8*). The relation $\gamma_{Kq} = \gamma_{qK}$ is due to the detailed balance condition satisfied by $\Sigma_s(E \to E')$.

In this formalism the γ_{Kq} are the characteristic parameters of the thermalization process. In the case of $1/v$ absorption we have $\Sigma_a(E) = \lambda_a/v$. We obtain

$$\lambda = \lambda_a + \frac{2v_0}{\sqrt{\pi}} D_{00} B^2 + \Delta \tag{7}$$

Δ can be written as a power series in B^2 starting with a term containing B^4. If we substitute (7) in the determinant $|\, d_{Kq}\,| = 0$ and retain terms up to B^4 only (*9*) we get a linear expression for Δ (up to B^4). If we assume that the off-diagonal elements in the matrix γ are small compared to the diagonal ones (as in the heavy gas model), then

$$\lambda = \lambda_a + \frac{2v_0}{\sqrt{\pi}} B^2 \left[D_{00} - B^2 \sum_{K=1}^{L} \frac{t_{0K}^2}{\gamma_{KK}}\right] \tag{8}$$

where

$$t_{0K} = D_{00}\, w_{0K} - D_{0K}$$

$$w_{0K} = \frac{2}{\sqrt{\pi}}\left(\frac{\sqrt{E}}{\sqrt{T}}\right)_{0K}$$

If $D(E) \sim 1/\sqrt{E}$ then $t_{0K} = 0$ and the leakage does not distort the spectrum.

One should be careful in analyzing thermalization data derived from "diffusion cooling" measurements for the

following reasons:

1. The t_{0K} (or D_{0K}) are not known to sufficient accuracy because $D(E)$ is not a directly measured quantity (*10*).

2. In order to obtain the nonlinearity in the $\lambda(B^2)$ curve one has to use very small systems where the accuracy of the diffusion approximation is questionable. Recently (*11*) it has been shown that by changing the shape of the system the coefficient of B^4 in λ can be changed by a factor of 2.

3. It is not possible by this method to derive more than one thermalization parameter without assuming certain simplifications.

We propose here another experimental method for measuring thermalization parameters like the γ_{Kq}. Assuming we add to an infinite moderator with "$1/v$" absorption a small concentration (N atoms/cm^3) of "non $1/v$" absorber with microscopic absorption cross section $\sigma_a(E)$. Then the spectrum of thermal neutrons after a short pulse will no longer be Maxwellian. The decay constant of the thermalized neutron flux will be

$$\lambda = \lambda_a + N\frac{\bar{\sigma}_a}{(1/v)} = \lambda_a + \frac{2v_0}{\sqrt{\pi}}N(\alpha + \beta N + \cdots) \tag{9}$$

where β is a measure of the distortion of the spectrum by the "non $1/v$" absorber. The transport equation for this case is (1) where B^2 is replaced by N and $D(E)$ by $\sigma_a(E)$. If $\sigma_a(E)$ in the thermal region is much larger than the absorption cross section of the moderator, then N may be very small compared to the density of the moderator atoms and yet the change of λ is relatively strong. In this case the value of λ is given by (8) where the t_{0K} are replaced by

$$s_{0K} = (\sigma_a)_{00}w_{0K} - (\sigma_a)_{0K}\,.$$

We have now to relate the above α and β to the values measured in a finite medium. Let us write for the decay constant in a finite system with added absorber

$$\lambda = \lambda' + \frac{2v_0}{\sqrt{\pi}}N(\alpha' + \beta' N + \cdots) \tag{10}$$

where λ' is the decay constant for the finite system of the pure moderator. Also take

$$\phi(E) = M_T(E)\sum_{K=0}^{L}(C_K^{(0)} + NC_K^{(1)} + N^2C_K^{(2)} + \cdots)L_K^{(1)}\left(\frac{E}{T}\right) \tag{11}$$

and we look at the various powers of N in the equation

$$\left[\frac{\lambda_a - \lambda}{v} + B^2D(E) + N\sigma_a(E)\right]\phi(E) = \int_0^\infty dE'[\Sigma_s(E' \to E)\phi(E') - \Sigma_s(E \to E')\phi(E)] \tag{12}$$

For the zeroth power of N we obtain (3) and λ' is given as before. From the linear term in N we obtain

$$\alpha' = \frac{\sum_{i,j=0}^{L} C_i^{(0)}C_j^{(0)}(\sigma_a)_{ij}}{\sum_{i,j=0}^{L} C_i^{(0)}C_j^{(0)}w_{ij}} \tag{13}$$

which in the case of $\gamma_{Kq} \ll \gamma_{KK}$ is (up to B^2)

$$\alpha' = (\sigma_a)_{00} - 2B^2\sum_{K=1}^{L}\frac{s_{0K}\,t_{0K}}{\gamma_{KK}} \tag{14}$$

From the N^2 term we obtain

$$\beta' = \frac{\sum_{i,j=0}^{L}((\sigma_a)_{ij} - \alpha' w_{ij})C_i^{(1)}C_j^{(0)}}{\sum_{i,j=0}^{L} C_i^{(0)}C_j^{(0)}w_{ij}} \tag{15}$$

For $L = 1$ it gives

$$\beta' = -\frac{s_{01}^2}{\gamma_{11}}\left(1 - \frac{B^2t_{01}}{\gamma_{11}}\left(w_{01} - \frac{s_{01}}{s_{11}}\right)\right) \tag{16}$$

These results are easily generalized for $L > 1$ and for γ_{Kq} comparable to γ_{KK} .

Experimentally, one measures the decay constant λ as a function of N, the concentration of the added absorber, and one gets $\alpha'(B^2)$ and $\beta'(B^2)$. The γ_{Kq} should be derived from β by the analogue to Eq. (8). The derivation of β from the measured $\beta'(B^2)$ can be performed in two ways: (i) measuring $\beta'(B^2)$ for several values of B^2 and extrapolating to $B^2 = 0$, and (ii) using Eqs. (13) and (15). It is preferred to use both methods. The extrapolation of $\alpha'(B^2)$ to $B^2 = 0$ should give $(\sigma_a)_{00}$, and this is used as a check.

When we compare this method to the "diffusion cooling" measurements we note the following:

1. Unlike t_{0K} , the s_{0K} may be readily calculated because $\sigma_a(E)$ is directly measured.

2. In general, measurements are performed in large enough systems so that the transport corrections to the diffusion approximation can be neglected.

3. By this method we separate the spectrum distorting mechanism from the moderator itself, and we can perform independent experiments on the same moderator with different absorbers. If it could be possible to describe a number of independent measurements by a smaller number of parameters, then one should compare these parameters to theory of $\Sigma_s(E \to E')$.

To this end we would like to have several "non $1/v$" strong absorbers in the thermal region. *Sm*, *Cd*, and *Gd* seem the only suitable ones. We notice that *Sm* has a resonance above K_BT (K_B is the Boltzman constant) while *Gd* has one below K_BT (for reasonable temperatures) and so their $(\sigma_a)_{01}$ have different signs. Adding 1–5% of *Gd* to *Sm* enables us to obtain s_{0K} over a wide range. In this way we can perform a large number of independent experiments. We see that the small number of available absorbing elements does not appear to be a serious disadvantage.

The above method is applicable in the present form to homogeneous systems only (e.g. solutions), but this semi-analytical approach may be a starting point to study thermalization in heterogeneous systems (*12*). Santandrea et al. (*13*) measured the dependence of λ on the concentration of *Cd* atoms in water for several values of B^2 and interpreted their results in terms of the dependence of "neutron temperature" on B^2. They gave $\beta'(B^2)$ with very poor accuracy.

Meadows and Whalen (*14*) observed the dependence of λ on N for "non $1/v$" absorbers while measuring the average over the Maxwellian of σ_a by the pulsed source technique in water solutions. They give β' for *Cd* and *Sm*, and with a large relative error for *Gd*, but this is not enough for our

purpose. It seems from their results that the single parameter γ_{11} is not sufficient to describe the process in water.

The experimental accuracy in measuring $\beta'(B^2)$ can be 1–2%. The s_{01} and s_{02} can be calculated with a relative error of about 1–2%, and the contribution of the other s_{0K} is relatively small. The γ_{Kq} for a certain moderator will be derived by a least square fitting of a maximum number of β to a minimum number of γ_{Kq} through the analogue to Eq. (8).

Experiments to obtain thermalization parameters by the above method are in progress in this laboratory.

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