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Measurements of Neutron Thermalization Parameters in Light Water by Pulsed Source and Non $1/v$ Absorber Method

Kiminori SHIBA*

Received June 28, 1967

Measurements of thermalization parameters in light water have been made by the pulsed neutron source and non $1/v$ absorber method. In an effectively infinite system of poisoned water, an asymptotic decay constant was measured by detecting capture γ-rays emitted from the whole volume of this system. The thermalization parameters were derived from values of decay constants measured over a wide range of non $1/v$ absorber concentration. Measured values of these parameters were compared with values calculated from Nelkin, Egelstaff-Schofield and McMurry-Russell kernels.

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I. INTRODUCTION

Neutron thermalization parameters in light water have been measured in an effectively infinite system, using the pulsed neutron source and non $1/v$ absorber method (Friedman's method). The asymptotic decay constant for the neutron density observed by this method is related to the concentration of the non $1/v$ absorber by the equation,

$$\lambda=\lambda_0+\alpha v_0 N-\beta v_0 N^2, \qquad (1)$$

where λ is the decay constant for the neutron density, N the number density of the non $1/v$ absorber atom, λ_0 the decay constant in pure light water, and v_0 the neutron velocity corresponding to an energy of K_BT, respectively (K_B being the Boltzmann constant, and T the absolute temperature of the system). Moreover, in Eq.(1), α is the effective absorption cross section, and β the absorption cooling or heating coefficient, whichever applicable according to whether the asymptotic neutron spectrum pertaining to the moderator system is cooled or heated by the non $1/v$ absorber. The coefficient β has the same physical meaning as the diffusion cooling coefficient obtained from an ordinary pulsed neutron experiment.

The advantages of the non $1/v$ absorber method have been pointed out by Friedman[1]. This method has consequently been applied to finite systems of water by several authors[2][3] and the values of absorption cooling (heating) coefficient have been reported for Cd, Sm and Gd. These values however have not shown agreement with the theoretical values calculated from the Nelkin kernel for water[4], despite consistence in the experimental diffusion cooling coefficient of water with the theoretical value derived from this kernel[5]. One of the causes of this discrepancy may be traced to the application of this method to finite systems of water. If this is the case, the experimental values should contain errors due to the higher spatial mode in the determination of the decay constant, as well as errors in the extrapolation to zero buckling, as suggested by Beckurts[5].

These errors can be eliminated by conducting experiments in which a decay of the total neutron number is observed in an effectively infinite system of water. In this case, the effect of neutron diffusion is suppressed. It is, however, laborious and unpractical to measure the decay of the total neutron number by ordinary neutron detectors such as BF_3 or ^3He counters, since these counters can detect the neutron density only at the point where it is placed, and the total number can not be obtained without numerical integration of the neutron density measured at many points. In order to overcome this difficulty, an attempt has been made to detect neutrons by the γ-rays emitted from the radiative capture of neutrons by nuclei. The same approach has been already tried in a large

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water assembly by others[6][7] who have successfully performed measurements of the thermal neutron absorption cross section. The high penetration power of γ-rays permits gaining information on neutrons in most parts of the system.

Utilizing this technique, the asymptotic decay constants were measured in an effectively infinite poisoned water system, and the values of absorption cooling (heating) coefficients have accurately been determined for Cd, Sm and Gd. The measured values of these coefficients were compared with theoretical calculations on the Nelkin[8], Egelstaff-Schofield[9] and McMurry-Russell kernels[10]. The calculations on these kernels were easier than by Calame's method[4][11], because of the condition adopted that the spectra perturbed by non $1/v$ absorbers had no net neutron density[12].

II. Theory

In the diffusion approximation, the neutron flux $\phi(\boldsymbol{r}, E, t)$ in a bare homogeneous moderator is represented by

$$\frac{1}{v}\frac{\partial}{\partial t}\phi(\boldsymbol{r}, E, t)=D(E)\nabla^2\phi(\boldsymbol{r}, E, t)-\Sigma_a(E)\phi(\boldsymbol{r}, E, t)-\Sigma_s(E)\phi(\boldsymbol{r}, E, t)+\int_0^\infty\Sigma_s(E'\to E)\phi(\boldsymbol{r}, E', t)dE', \quad (2)$$

where $D(E)$ is the diffusion coefficient, $\Sigma_a(E)$ the macroscopic absorption cross section, $\Sigma_s(E)$ the macroscopic total scattering cross section, and $\Sigma_s(E'\to E)$ the scattering kernel. When a γ-ray detector is placed in a neutron field, the count of the detector $C(t)$ is related to the neutron flux by

$$C(t)=\int d\boldsymbol{r}\int_0^\infty dE\int_0^\infty dE_\gamma\int_0^\infty dE_\gamma'\Sigma_d(E, E_\gamma)\cdot\phi(\boldsymbol{r}, E, t)F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')P(E_\gamma'), \quad (3)$$

where $\Sigma_d(E, E_\gamma)$ is the macroscopic cross section of radiative capture in which γ-rays of energy E_γ are emitted from a nucleus absorbing a neutron of energy E, $F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')$ is the probability that γ-rays of energy E_γ emitted at $\boldsymbol{r}$ will arrive at $\boldsymbol{r}_0$ (where the γ-ray detector is placed) with energy E_γ', and $P(E_\gamma')$ is the efficiency of the γ-ray detector. Hence the count of the detector $C(t)$ can be directly combined with the integrated flux $\bar{\phi}(E, t; \boldsymbol{r}_0)$ defined by

$$\bar{\phi}(E, t; \boldsymbol{r}_0)=\int d\boldsymbol{r}\int_0^\infty dE_\gamma\int_0^\infty dE_\gamma'\Sigma_d(E, E_\gamma)\phi(\boldsymbol{r}, E, t)\cdot F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')P(E_\gamma'). \quad (4)$$

In order to obtain an equation for $\bar{\phi}(E, t; \boldsymbol{r}_0)$, Eq.(2) is multiplied by $\Sigma_d(E, E_\gamma)F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')P(E_\gamma')$ and integrated with respect to $\boldsymbol{r}$, E_γ and E_γ'. Thus the equation

$$\frac{1}{v}\frac{\partial}{\partial t}\bar{\phi}(E, t; \boldsymbol{r}_0)=D(E)\mu_0(E, t; \boldsymbol{r}_0)\bar{\phi}(E, t; \boldsymbol{r}_0)-\Sigma_a(E)\bar{\phi}(E, t; \boldsymbol{r}_0)-\Sigma_s(E)\bar{\phi}(E, t; \boldsymbol{r}_0)+\int_0^\infty\Sigma_s(E'\to E)\bar{\phi}(E, t; \boldsymbol{r}_0)dE' \quad (5)$$

is obtained, where

$$\mu_0(E, t; \boldsymbol{r})=\frac{\int d\boldsymbol{r}\int_0^\infty dE_\gamma\int_0^\infty dE_\gamma'\nabla^2\phi(\boldsymbol{r}, E, t)\cdot\Sigma_d(E, E_\gamma)F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')P(E_\gamma')}{\int d\boldsymbol{r}\int_0^\infty dE_\gamma\int_0^\infty dE_\gamma'\phi(\boldsymbol{r}, E, t)\cdot\Sigma_d(E, E_\gamma)F(\boldsymbol{r}, E_\gamma|\boldsymbol{r}_0, E_\gamma')P(E_\gamma')} \quad (6)$$

When a large moderator system is used, and a detector detects γ-rays emitted from most part of the system, the relation $|D(E)\mu_0(E, t; \boldsymbol{r}_0)/\Sigma_a(E)|\ll 1$ can be expected to hold. And so the term containing $\mu_0(E, t; \boldsymbol{r}_0)$ is neglected, and consequently the coordinate of the detector position $\boldsymbol{r}_0$ can be omitted. Then Eq.(5) reduces to

$$\frac{1}{v}\frac{\partial}{\partial t}\bar{\phi}(E, t)=-\Sigma_a(E)\bar{\phi}(E, t)-\Sigma_s(E)\bar{\phi}(E, t)+\int_0^\infty\Sigma_s(E'\to E)\bar{\phi}(E', t)dE'. \quad (7)$$

Since this equation contains no term representing the effect of the diffusive motion of neutrons, diffusion does not contribute to the asymptotic decay of the integrated flux $\bar{\phi}(E, t)$. Here we divide the absorption of neutrons in the system into two parts: (1) the absorption by the moderator itself, and (2) that by the non $1/v$ absorber. Then the asymptotic decay constant λ obtained from the observation of $\bar{\phi}(E, t)$ is the smallest eigenvalue of the equation

$$(\lambda/\Sigma_{s0}v-\Sigma_{a0}v_0/\Sigma_{s0}v)\bar{\phi}(E)=\boldsymbol{H}\bar{\phi}(E)/\Sigma_{s0}+\varepsilon a(E)\bar{\phi}(E), \quad (8)$$

where

$$\boldsymbol{H}\bar{\phi}(E)=\Sigma_s(E)\bar{\phi}(E)-\int_0^\infty\Sigma_s(E'\to E)\bar{\phi}(E')dE', \quad (9)$$

$$a(E)=\Sigma_a^*(E)/\Sigma_{a0}^*, \quad (10)$$

$$\varepsilon=\Sigma_{a0}^*/\Sigma_{s0}. \quad (11)$$

Moreover, in Eq.(8), the absorption cross section of an unpoisoned moderator is assumed to obey the $1/v$ law, and described as $\Sigma_{a0}v_0/v$ (Σ_{a0} being the macroscopic absorption cross section of moderator at $v=v_0$), Σ_{s0} is the macroscopic scattering cross section of free atom of the moderator, $\Sigma_a^*(E)$ the macroscopic absorption cross section of non $1/v$ absorber, and Σ_{a0}^* the macroscopic absorption cross section of non $1/v$ absorber at $v=v_0$.

Considering that $\varepsilon a(E)\bar{\phi}(E)$ is a perturbed term, we will solve Eq.(8) by the perturbation technique[4]. Then λ and $\bar{\phi}(E)$ are expanded into power series:

$$\lambda=\lambda_0+\varepsilon\lambda_1-\varepsilon^2\lambda_2+\cdots\cdots, \quad (12)$$

$$\bar{\phi}(E)=\phi_0(E)-\varepsilon\phi_1(E)+\cdots\cdots. \quad (13)$$

Substituting Eqs.(12) and (13) into Eq.(8), and equating the coefficients of the terms of equal ε^n yields a system of equations that can be solved successively, beginning with the $n=0$ equation:

$$(\lambda_0-\Sigma_{a0}v_0)\phi_0(E)/v=\boldsymbol{H}\phi_0(E). \quad (14)$$

The operator $\boldsymbol{H}$, which appears in Eq.(14) and also in the subsequent equations, satisfies the neutron conservation condition, signifying vanishment of the integration of $\boldsymbol{H}\phi_0(E)$ over all energies. Thus the solution of Eq.(14) is the well known Maxwellian flux spectrum $M(E)$ associated with the eigenvalue $\lambda_0=\Sigma_{a0}v_0$.

The values of λ_1 can be obtained by integrating the $n=1$ equation

$$\lambda_1 M(E)/\Sigma_{s0}v=-\boldsymbol{H}\phi_1(E)/\Sigma_{s0}+a(E)M(E) \quad (15)$$

over all energies. This procedure gives the result

$$\lambda_1=\Sigma_{s0}\int_0^\infty a(E)M(E)dE\Big/\int_0^\infty M(E)/vdE$$

$$=\frac{2}{\sqrt{\pi}}\Sigma_{s0}v_0\int_0^\infty a(E)M(E)dE. \quad (16)$$

With λ_1 known, Eq.(15) may be solved for $\phi_1(E)$.

Then λ_2 is found by integrating over energy the $n=2$ equation

$$\lambda_2 M(E)/\Sigma_{s0}v+\lambda_1\phi_1(E)/\Sigma_{s0}v$$

$$=-\boldsymbol{H}\phi_2(E)/\Sigma_{s0}+a(E)\phi_1(E), \quad (17)$$

the result being

$$\lambda_2=\Sigma_{s0}\int_0^\infty[a(E)-\lambda_1/\Sigma_{s0}v]\phi_1(E)dE\Big/\int_0^\infty M(E)/vdE$$

$$=\frac{2}{\sqrt{\pi}}\Sigma_{s0}v_0\int_0^\infty[a(E)-\lambda_1/\Sigma_{s0}v]\phi_1(E)dE. \quad (18)$$

Since it is known that a perturbed flux $\phi_1(E)$ does not contribute to the net neutron density[12], *i.e.*,

$$\int_0^\infty\phi_1(E)/vdE=0, \quad (19)$$

Eq.(18) reduces to

$$\lambda_2=\frac{2}{\sqrt{\pi}}\Sigma_{s0}v_0\int_0^\infty a(E)\phi_1(E)dE. \quad (20)$$

Equation (19) will be utilized in Chap. Ⅳ to solve Eq.(15) numerically for $\phi_1(E)$.

Since $\Sigma_a^*(E)$ can be written $N\sigma_a^*(E)$ using the number density N and the microscopic absorption cross section $\sigma_a^*(E)$ of the non $1/v$ absorber, the decay constant is expressed in the same form as Eq.(1). Coefficients α and β, related to λ_1 and λ_2 by $\alpha=\lambda_1\varepsilon/v_0N$ and $\beta=\lambda_2\varepsilon^2/v_0N^2$, are the effective absorption cross section and the absorption cooling (heating) coefficient, respectively.

Ⅲ. Experimental Method

The moderator system used was poisoned water in a cubic container, $72\times72\times75\,cm^3$, made of lucite plates 7 mm thick. The neutron source was placed at the center of one of the walls of this container. The γ-rays emitted from neutron capture by atoms were detected by an NaI crystal 2″ diameter and 2″ long. The crystal was positioned on the central axis of the moderator system, and arranged to move along the axis; it was connected to a DuMont 6292 photomultiplier* through a light pipe (50 mm dia.×350 mm long) of lucite.

A Cockcroft-Walton type neutron generator was used as pulsed neutron source[13]. The pulsed deuteron beam was produced by means of a grid plate to which pulsated voltage was applied, and which bombarded a

* It has been pointed out that the DuMont 6292 photomultiplier changes pulse height with counting rate. In the case of the tube used in this work, it was ascertained that this change in pulse height was only a few % for a factor of 100 in counting rate. Experimental data were therefore not corrected for this effect.

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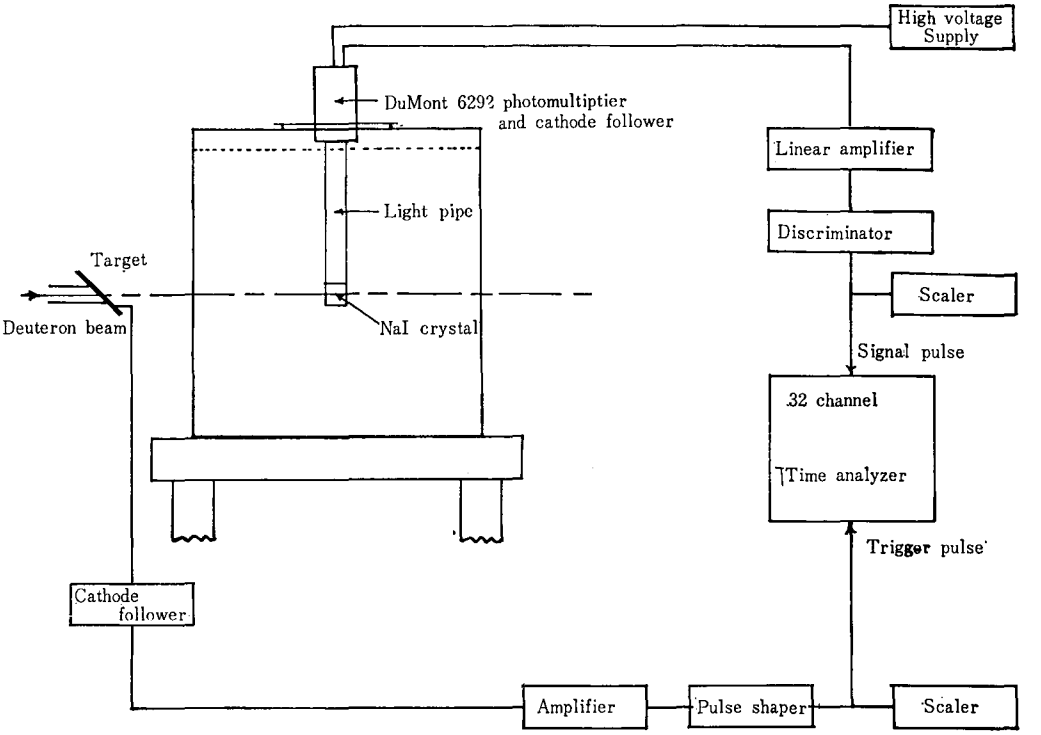


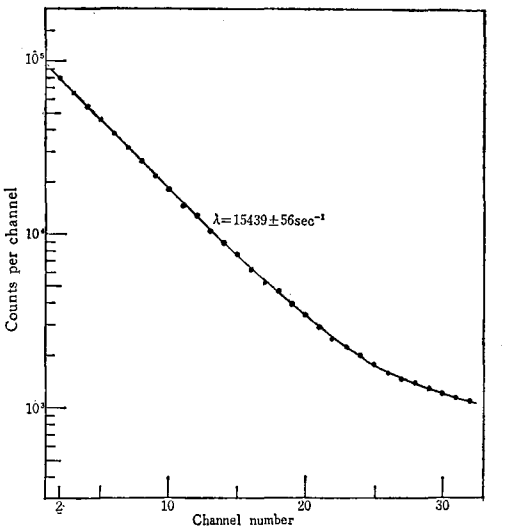
Fig. 1 Measuring system

tritium target. The pulse widths were chosen to be 25, 10 and 5 μsec, with a repitition rate of 100 Hz, 300 Hz and 1 kHz, respectively. The accelerating voltage used was from 250 to 280 kV.

The signals from the γ-ray detector were fed into a voltage discriminator after amplification, and only signals produced by γ-rays with energy above 2.2 MeV were selected. The out-put signals from the discriminator were fed into a 32-channel time-analyzer[14], the zero time signal there of being obtained from the target struck by pulsed beam. Channel width used was switched from 25 to 12.5 and to 6.25 μsec, as the decaying rate increased.

The measuring system is shown in **Fig. 1** and an example of observed decay curve **Fig. 2.** The data of counts *vs.* time were fitted to $A+B\exp(-\lambda t)$ by least squares.

Owing to the dead time of the circuit system and a surviving higher mode of energy, the earlier part of the decay curves has a form that is not exponential. Thus, the values of decay constant determined by least squares varied with the start channel, and gradually approached fixed values. These



Channel width: 12.5 μsec
Non $1/v$ absorber: Sm
Concentration: 5.50×10^{18} Sm-atoms/cm^3
(Background not subtracted)

Fig. 2 Typical decay curve

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final values can therefore be considered unaffected either by the dead time or by the surviving higher mode of energy.

The number density of the added absorber was determinded by chelate titration using EDTA reagent[15]. Cd, Sm and Gd were used as non $1/v$ absorber. The maximum number densities of the non $1/v$ absorbers used in this experiment were 2.2×10^{19}, 1.4×10^{19} and 3.6×10^{18} at./cm^3 for Cd, Sm and Gd respectively. These values correspond to $\varepsilon\leqq0.1$, as defined by Eq.(11), so that the perturbation technique can be applied to the analysis of this experiment.

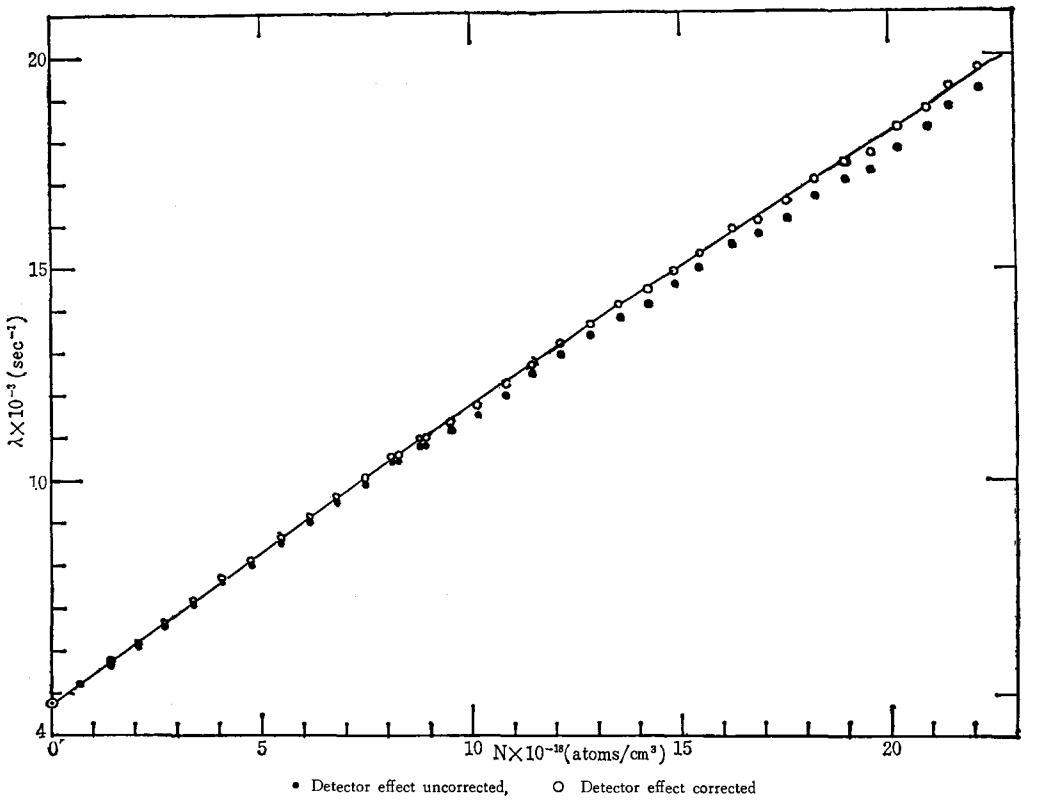
IV. Experimental Results

In order to apply the theory described in Chap. II to the analysis of the experimental data, the assumption, $|D(E)\mu_0(E, t; r_0)/\Sigma_a(E)| \ll 1$, must be confirmed for this measuring system. To this end, the decay constants of neutron density in pure water were measured for several positions of the detector, and compared with the values of $\Sigma_{a0}v_0$ obtained from the ordinary pulsed experiment[16] and with those from measurements of diffusion length in a non-stationary neutron field[17]. The values agreed with each other as shown in **Table 1**, and the above assumption was thus considered applicable under experimental conditions.

Table 1 Comparison of decay constants in pure water system with values of $\Sigma_{a0}v_0$ obtained from other experiments

Decay constant (sec^{-1})			$\Sigma_{a0}v_0$ (sec^{-1})	
Detector position†			Author	
18 cm	36 cm	54 cm	Lopez[16] & Beyster	Arai[17] & Küchle
4,793±25	4,747±24	4,728±26	4,768±24	4,782±15

† Detector positions indicated in terms of the distance between detector and container wall facing neutron source.



• Detector effect uncorrected, ○ Detector effect corrected

Fig. 3 Decay constants λ *vs.* non $1/v$ absorber number density N in Cd poisoned water

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Because the space occupied by the γ-ray detector cannot be poisoned with non $1/v$ absorbers, the mean concentration of the added absorber is thereby lowered around the detector. Thus, the measured decay constant is influenced by the presence of the detector itself. In order to estimate this effect, the decay constants were measured in a system containing an additional unpoisoned space having the same volume as the γ-ray detector, and the results were compared with the decay constants in the system without this additional space. The additional space was produced by a cylindrical lucite container filled with pure water, which was placed adjacent to the γ-ray detector. This measurement was made in a system poisoned with Sm. Here it was assumed that this additional space should provide the same effect on the decay constant as the γ-ray detector.

It was thus defined that the true decay constant λ in the homogeneously poisoned system could be combined with the decay constant λ' observed in the system containing the γ-ray detector by the relation

$$\lambda-\lambda_0=(1.035\pm0.004)(\lambda'-\lambda_0), \qquad (21)$$

where λ_0 is the decay constant in a pure water system. Then the measured values of decay constants can be corrected with the use of Eq.(21). The values of decay constants thus corrected plotted against the number densities of Cd, Sm and Gd atoms in **Figs. 3, 4** and **5** respectively.

The parameters α and β of Eq.(1) have been calculated by least squares fitting of these curves. The effective absorption cross sections α thus obtained are given in **Table 2**, where these values are compared with the values calculated with the use of cross section values from *BNL*-325 and of the resonance parameters given by Westcott[18]. Numerical integration of Eq.(16) was carried out over the energy range 0~1 eV divided into 300 mesh. It is seen from Table 2 that the calculated values agree well with experimental data, which provides evidence in support of the correction adopted for the decay constant to account for the presence of the γ-ray detector, as expressed by Eq.(21).

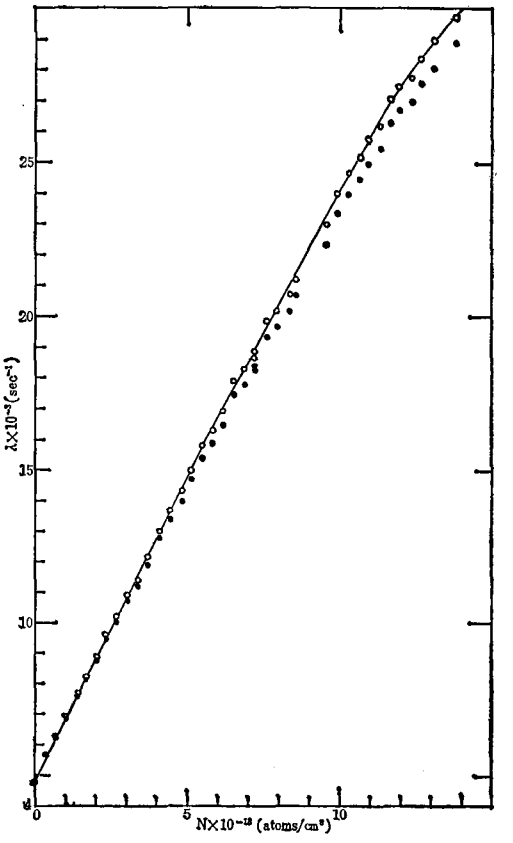

Fig. 4 Decay constant λ *vs.* non $1/v$ absorber number density N in Sm poisoned water

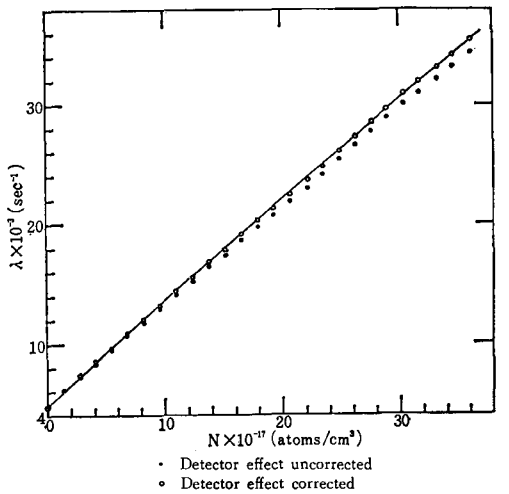

Fig. 5 Decay constant λ *vs.* non $1/v$ absorber number density N in Gd poisoned water

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Table 2 Effective absorption cross section

Non $1/v$ absorber	Water temp. (°C)	Effective absorption cross section (barns)		Calculation / Experiment
		Experiment	Calculation	
Cd	15±2	3,340±15	3,277	0.98
Sm	27.6±1.3	9,360±48	9,455	1.01
Gd	21.5±0.7	40,400 ±180	40,950	1.01

In **Table 3**, it is seen that the absorption cooling coefficients of Cd and Sm are smaller than the values by Friedman[2], and by Meadow & Whalen[3], by about 20% and 30%, respectively, however for Gd our absorption heating coefficient is about twice that obtained by Meadow & Whalen.

Table 3 Experimental absorption cooling (heating) coefficients (10^{-17} barns·cm^3)

Non $1/v$ absorber	Present	Friedman†	Meadow & Whalen
Cd	1.40±0.08 (15±2°C)	1.75±0.28 (23°C)	1.75±0.07 (25°C)
Sm	7.00±0.45 (27.6±1.3°C)	9.5±2.4 (23°C)	9.73±0.12 (25°C)
Gd	53.5±4.8 (21.5±0.7°C)	—	26±13 (25°C)

† Values in reference multiplied by $2/\sqrt{\pi}$.

V. Calculation of Absorption Cooling (Heating) Coefficients

In order to calculate the absorption cooling (heating) coefficient from Eq.(20), the perturbed flux $\phi_1(E)$ must be known. The Maxwellian flux satisfies the homogeneous equation $\boldsymbol{HM}(E)=0$, so that an additional condition is necessary to obtain a unique solution. This additional condition can be considered adequately provided by Eq.(19). Thereupon, Eq.(15) was transformed into a system of 30 linear equations. In this case the energy range 0~0.9 eV was divided into 30 sections having equal or nearly equal velocity intervals. With the use of Eq.(19), expressed in the form of a summation over the divided regions, the perturbed flux $\phi_1(E)$ was calculated numerically from Eq.(15). Calculation by this means is easier than with Calame's method[4][11] which requires a laborious iterative operation.

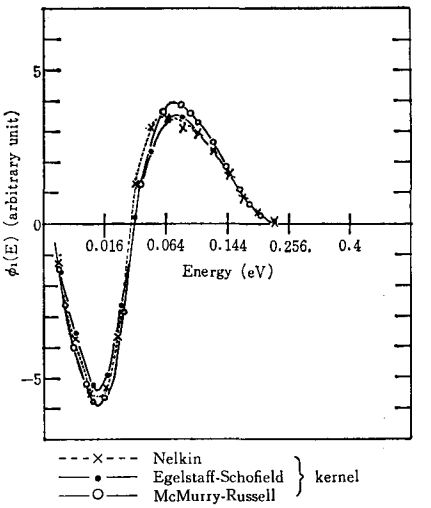

Fig. 6 Flux $\phi_1(E)$ perturbed by Cd

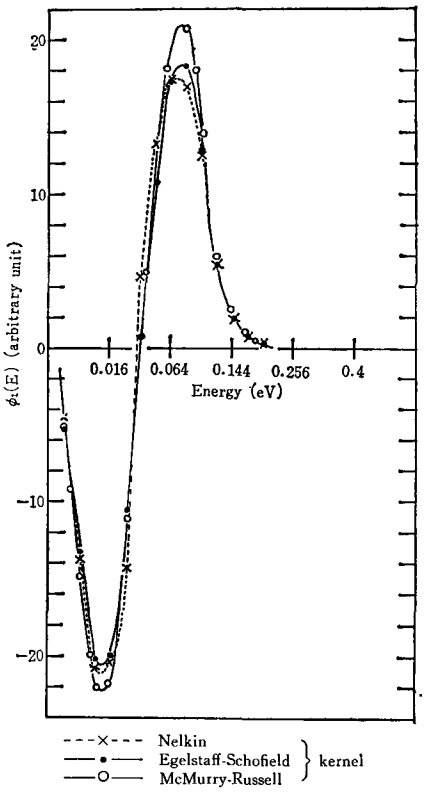

Fig. 7 Flux $\phi_1(E)$ perturbed by Sm

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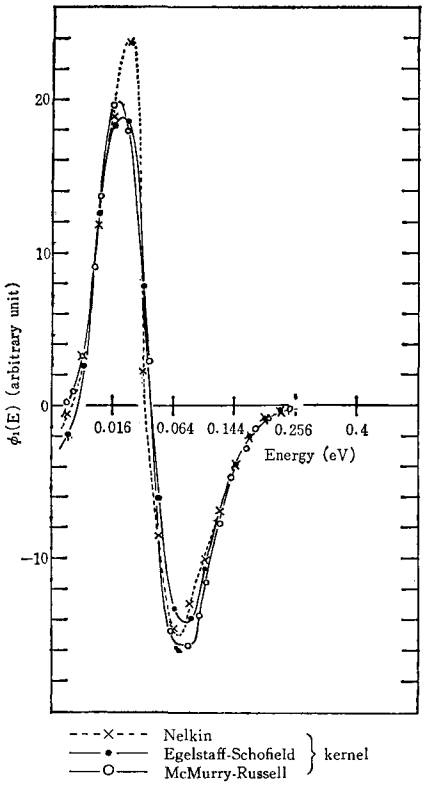

Fig. 8 Flux $\phi_1(E)$ perturbed by Gd

In calculating the perturbed flux, the Nelkin[8], Egelstaff-Schofield[9] and McMurry-Russell kernels[10] were used. The perturbed fluxes by non $1/v$ absorbers are shown in **Figs. 6, 7** and **8**, and the values of absorption cooling (heating) coefficients calculated from Eq.(20) given in **Table 4**. These values are nearly independent of the kernels used in this calculation and are in agreement with those by Calame[4] for the Nelkin kernel. There is however wide disagreement between the experimental and calculated values.

Table 4 Calculated absorption cooling (heating) coefficients (10^{-17} barns·cm^3)

Non $1/v$ absorber	Present			Calame[4]
	Nelkin kernel[8] (20°C)	Egelstaff-Schofield kernel[9] (20°C)	McMurry-Russell kernel[10] (22°C)	Nelkin kernel[8] (25°C)
Cd	0.330	0.325	0.362	0.329
Sm	3.50	3.55	3.98	3.48
Gd	3.13	3.18	3.39	4.18

VI. Summary

Measurements of the decay constant of the neutron density in an effectively infinite poisoned water system have been carried out, with the use of γ-ray detector. Accurate determination of the effective absorption cross section and of the absorption cooling (heating) coefficients for Cd, Sm and Gd have been made from the experimental values of decay constant over a wide range of the non $1/v$ absorber concentration. The values of the effective absorption cross section agreed well with calculation.

The experimental values of absorption cooling coefficients of Cd and Sm are smaller than those by Friedman[2], and by Meadow & Whalen[3], but the absorption heating coefficient of Gd turns out to be about twice as large as Meadow-Whalen's. The present values differ considerably from all those calculated on the basis of the Nelkin[8], Egelstaff-Schofield[9] and McMurry-Russell[10] kernels.

The condition that a flux perturbed by non $1/v$ absorber does not contribute to the net neutron density[12], make the calculation of absorption cooling (heating) coefficients much easier than by Calame's method[4][11].

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