PULSED NEUTRON MEASUREMENTS OF THE DIFFUSION PARAMETERS OF HEAVY AND LIGHT WATER MIXTURES AT SEVERAL TEMPERATURES*

by

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TABLE OF CONTENTS

LIST OF TABLES

LIST OF FIGURES

I. INTRODUCTION

Knowledge of the diffusion parameters for thermal neutrons in a moderating material is essential for reactor design. The diffusion coefficient, or the related quantity the transport mean free path, is used to determine the over-all leakage from a reactor.

The proposed use of mixtures of light and heavy water as moderator in power reactors has suggested an experimental determination of the diffusion parameters of such mixtures by the pulsed neutron technique. As the diffusing neutrons are in thermal contact with the moderating medium, one should expect a temperature dependence of the diffusion parameters. Clendenin (1) has shown that measurements of the temperature dependence of these parameters may provide a method for descriminating between different theoretical scattering models.

Experimental data are available on the neutron diffusion parameters of light and heavy waters at room and elevated temperatures. Very little experimental data are available on the parameters for light water ice and no published results have been noted for light-heavy water mixtures or for heavy water at temperatures below 10°C.

The experiments described were undertaken to determine the diffusion parameters of 0, 20, 50, 80 and 100 per cent (by volume) heavy water near the freezing temperature in the liquid state and at -20°C in the solid state. Similar

measurements on the above mixtures have been made at room temperature (2).

The method consists of injecting bursts of neutrons from the $_1H^2(d,n)_2He^3$ reaction into different sizes of the moderator medium and recording the exponential decay of the neutron counting rate with a thermal neutron detector external to the medium.

Decay constants, λ, were obtained for different bucklings and temperatures with a multi-channel time analyzer. Diffusion parameters, D_o and C, were computed by a least-squares fit of the λ vs B^2 data to the expression:

$$\lambda = \lambda_o + D_o B^2 - CB^4$$

II. LITERATURE REVIEW

The pulsed neutron technique has been used extensively to determine neutron diffusion parameters of moderators, primarily by measurement of neutron mean lifetimes as a function of geometric size.

Light water has been investigated over a range of temperatures by Von Dardel and Sjostrand (3), A.V. Antonov et al. (4,5), Dio and Schopper (6), Bracci and Coceva (7), Lopez and Beyster (8), and Küchle (9).

Tables I and II summarize a number of measurements of light water at different temperatures. The measurements of Antonov et al. were in the temperature range of 0° to 286°C and on ice at -80° and -196°C. Clendenin (1) used both the Nelkin and Radkowsky models to calculate the diffusion coefficients for water in the temperature range of 23° to 300°C. Dlouhy and Kvitek (10) reported measurements on water at 20° and 0°C and on ice at 0°C. McClure (11) measured the parameters for water at 1.0°C and ice at -19°C. Very recently, Silver (12) made measurements on ice at -5°, -25°, -45°, -65° and -85°C.

The harmonically modulated source method was used by Raievski and Horowitz (13) to measure D_o in heavy water. Pulsed experiments in heavy water have been performed by Sjostrand (14), Ganguly et al. (15), Westfall and Waltner (16), and Kussmaul and Meister (17). Honeck and Michael (18)

reported theoretical values of D_o and C for heavy water at 20°, 77°, 127°, and 177°C. These results were obtained by solving the Boltzmann equation, using a theoretical scattering model previously proposed by Honeck (19).

Baumann (20) measured the diffusion coefficients for thermal neutrons in heavy water by a static method utilizing a copper-poisoning technique. Results were obtained at 20°, 100°, 165°, and 220°C.

One other set of experimental data in this temperature range was reported recently by Daughtry and Waltner (21). The pulsed neutron technique was used to obtain results in heavy water in the temperature range of 25° to 250°C.

Table III is a summary of diffusion parameter measurements for heavy water.

Jones (2) measured the diffusion parameters of 0, 20, 50, 80, and 100 percent (by volume) heavy water at room temperature for buckling values ranging from 0.058 cm^{-2} to 0.65 cm^{-2}.

Other than the above results there is no known data on neutron diffusion parameters for pure heavy water below 10°C or on light-heavy water mixtures below room temperature (22).

TABLE I

Diffusion Parameters for H_2O by the Pulsed Neutron Method

Reference	$D_o(cm^2sec^{-1})$	$C(cm^4sec^{-1})$	σ_a^H(mb)	L(cm)	B^2 range cm^{-2}	Temp (°C)
Manley et al. (23)	--	--	345	--	Large Geometry	22
Von Dardel and Waltner (24)	--	--	321± 5	--	Infinite Geom.	22
Scott et al. (25)	38500 ± 800	--	320± 8	2.85± 0.05	0.006- 0.018	22
Von Dardel and Sjostrand (3)	36340 ± 750	7300±1500	333 ± 3	2.725±0.03	0.1 - 0.7	22
Antonov et al. (4)	35000±1000	4000±1000	329±10	2.7 ± 0.1	0.09 - 0.93	22
Bracci and Coceva (7)	34850±1100	3000±1000	337±10	2.66 ± 0.11	0.09 - 0.96	22
Meads et al. (26)	--	--	335 ± 4	--	Infinite Geom.	22
Campbell and Stelson (27)	34800	0	327	2.69	0.08 - 1.1	22
Stooksberry and Marshall (28)	--	--	330 ± 8	--	Large Geom.	22
Dio (29)	35450 ± 600	3700 ± 700	328 ± 6	2.715±0.06	0.093 - 0.87	22
Kuchle (9)	35400 ± 700	4200 ± 800	326± 6	2.72 ±0.08	0.11 - 0.75	22
Lopez and Beyster (8)	37503 ± 366	5116 ± 776	325± 1.6	2.828±0.016	0.0137-0.591	26
	36892 ± 400	--	--	2.803±0.016	0.0137-0.591	22
Bretscher (30)	36985±1630	5021±2490	322±17	2.82 ± 0.09	0 - 0.60	26
Beckurts (31)	34820 ± 720	3650 ± 400		--		22
Antonov et al. (5)	35000±1000	4000±1000	329±10	2.7 ± 0.1	0.09 - 0.93	22
Dlouhy and Kvitek (10)	34600 ± 800	4000 ± 800	--	--		20
Kovel (32)	37405 ± 422	5162 ±2144	--	--	0.1085-0.248	24
McClure (11)	29670±2640	92±3550	--	--	0.150 - 0.790	0
Jones (2)	35800 ± 547	7094±1455	--	--	0.0586-0.6508	21

TABLE II

Diffusion Parameters for H_2O by Other Methods

Reference	D_o ($cm^2 sec^{-1}$)	C ($cm^4 sec^{-1}$)	σ_a^H (mb)	L (cm)	Method	Temp (°C)
DeJuren et al. (33)	--	--	--	2.763±0.015	DLM*	22
Hammermesh et al. (34)	--	--	329 ± 4	--	BPE**	22
Harris et al. (35)	--	--	332 ± 7	--	BPE	22
Barkow et al. (36)	--	--	--	2.71 ± .02	DLM	22
Baker (37)	--	--	327 ± 4	--	BPE	22
Reier et al. (38)	--	--	--	2.774 ± .007	DLM	22
Reier (39)	37618 ± 205	--	328 ± 6	--	BPE	22
Rockey et al. (40)	--	--	--	2.838 ± .018	DLM	22
Starr et al. (41)	35850 ± 100	2900 ± 350	326.9 ± 1.6	2.754 ± .008	DLM	21

*Diffusion Length Measurement

**Boron Poisoning Experiments

TABLE III

Diffusion Parameters for D_2O

Reference	$D_o x10^{-5}$ (cm^2/sec)	L(cm)	$Cx10^{-5}$ (cm^4/sec)	Temp. (°C)	λ_{tr} (cm)
Auger et al. (42)	--	--	--	22	2.4 ± 0.1
Sargent et al. (43)	--	171 ± 20	--	22	--
Kash & Woods (44)	--	--	--	20	2.52 ± 0.04
Raievski & Horowitz (13)	2.00 ± 0.05	--	--	13	2.45 ± 0.07
Sjostrand (14)	--	--	3.5 ± 0.8	20	--
Baumann (45)	--	--	--	20	2.52 ± 0.03
Starr & de Villiers (46)	--	--	5		--
Westfall & Waltner (16)	2.040 ± 0.013	--	4.19 ± 0.18	29	--
Ganguly et al. (15)	2.00 ± 0.04	--	2.95 ± 0.43	10	--
	2.08 ± 0.05	--	3.72 ± 0.50	20	--
Kussmaul & Meister (17)	2.0 ± 0.009	--	5.25 ± 0.25	22	--
Jones (2)	1.966 ± 0.013	--	3.56 ± 1.08	21	2.670
Malaviya & Profio (47)	2.045 ± 0.044	--	4.706 ± 0.381	21	--
Honeck & Michael (18)	2.057	--	4.73	20	--
	2.354	--	5.31	77	--
	2.658	--	6.05	127	--
	3.011	--	7.43	177	--
Daughtry & Waltner (21)	1.983 ± 0.029	--	4.65 ± 0.54	25	2.378
Parks & Baumann (49)	2.10 ± 0.02	--	8.1 ± 0.3	20	--

III. THEORY OF THE PULSED NEUTRON METHOD

The pulsed neutron method for measuring the diffusion parameters of moderators consists of irradiating the moderator sample by a short burst of fast neutrons and observing the decay of the neutron density at the surface of the moderator. Inside the moderator the fast neutrons slow down to thermal velocities. When the neutrons approach thermal equilibrium with the moderator, the neutron density may be approximated by the single group diffusion equation:

$$D\nabla^2 n - v\Sigma_a n = \frac{\partial n}{\partial t} \tag{1}$$

$$D = \frac{\lambda_{tr}}{3} v$$

where

n = neutron density

D = diffusion coefficient

λ_{tr} = transport mean free path

Σ_a = macroscopic absorption cross section

the solution which satisfies the condition that the flux vanishes at the extrapolated boundary is

$$n(\bar{r},t) = \sum_{lmn} A_{lmn} S_{lmn}(\bar{r}) e^{-(v\Sigma_a + DB^2_{lmn})t} \tag{2}$$

S_{lmn} and B^2_{lmn} are respectively the eignefunctions and eigenvalues of the Helmholtz equation:

$$(\nabla^2 + B^2_{lmn})\ S_{lmn} = 0$$

$S_{lmn}(\bar{r})$ are zero at the extrapolated boundaries of the medium. A_{lmn} are constants that depend on the initial distribution of the neutrons.

After a reasonable time the density of neutrons is determined by the fundamental mode of the summation, that is the term with the smallest decay constant becomes the dominant term. Thus, equation 2 becomes:

$$n(\bar{r},t) = ASe^{-\lambda t}$$

$$\lambda = v\Sigma_a + DB^2$$

where the indices l = m = n = 1 have been dropped for simplification. Equation 1 describes the diffusion of monoenergetic neutrons. Although the neutrons are not monoenergetic, the neutron density still satisfied equation 1 if the parameters are averaged over the flux. For neutrons diffusing through a finite size moderator, a two group approximation leads to a better qualitative understanding of the diffusion process. In this case the diffusion equations take the form:

$$(D_1\nabla^2 - \Sigma_a v - \beta_{12})n_1 + \beta_{21}n_2 = \frac{\partial n_1}{\partial t} \qquad (3)$$

$$(D_2\nabla^2 - \Sigma_a v - \beta_{21})n_2 + \beta_{12}n_1 = \frac{\partial n_2}{\partial t} \qquad (4)$$

D_1 and D_2 are the diffusion coefficients for neutrons of the first and second groups, β_{12} and β_{21} are the probabilities per unit time for neutrons of group n_1 to be scattered into group

n_2 and for neutrons of group n_2 to be scattered into group n_1.

For simplification, it will be assumed that the product $v\Sigma_a$ and the buckling B^2 are the same for neutrons of both groups, and that $\beta_{12} = \beta_{21} = \beta$. The solution of equations 3 and 4 is:

$$n = n_1+n_2 = e^{-(\Sigma_a v + \frac{D_1+D_2}{2} B^2+\beta)t}(Pe^{\mu t}+Qe^{-\mu t}) \qquad (5)$$

P and Q are constants and

$$\mu^2 = \beta^2 + \frac{(D_1 - D_2)^2}{8\beta} B^4$$

Equation 5 indicates that the neutron density can be described by the sum of two exponential terms with different decay periods. Suppose that after a certain time that equilibrium between the two groups is established, that is, the term with the larger decay constant can be neglected compared with the other term. This means that the difference in neutron leakage of both groups is small. In such a case μ can be expanded into a power series in B^2, leading to

$$n \simeq pe^{-\lambda t}$$

where

$$\lambda = \Sigma_a v + \frac{D_1+D_2}{2} B^2 - \frac{(D_1-D_2)^2}{8\beta} B^4 + \ldots \qquad (6)$$

$$= \Sigma_a v + D_o B^2 - CB^4$$

Where B^2 is the buckling of the sample, $\Sigma_a v = \lambda_o$, is the decay constant for an infinite medium ($B^2 = 0$), D_o is the average diffusion coefficient for thermal equilibrium, and C is a

positive parameter whose magnitude depends on the spread of D(E) over the neutron spectrum. Parameter C is called the diffusion cooling constant.

Equation 6 is based on the diffusion theory approximation including the diffusion cooling effect in a finite size moderator as a consequence of the preferential leakage of fast neutrons. In applying equation 6 to the interpretation of pulsed neutron experiments one assumes that a fundamental mode has been established and a single buckling may be assigned to the sample and that the neutron density vanishes at the extrapolated boundaries of the moderator.

For cylindrical geometry the buckling is given by:

$$B^2 = [2.405/(R + \varepsilon)]^2 + [\pi/(H + 2\varepsilon)]^2$$

ε is the extrapolation distance; R and H are the radius and height of the cylinder. The two group theory above was developed by Antonov et. al. (5). If the neutron spectrum were a Maxwellian at the moderator temperature, T, the second term to the right of equation 6 would correctly describe the effects of leakage on the decay constant, since D_o is the average of the energy dependent diffusion coefficient D(E):

$$D_o = \frac{\int_0^\infty D(E)M(E)dE}{\int_0^\infty \frac{1}{v} M(E)dE}$$

$$M(E) = \frac{E}{(kT)^2} e^{-E/kT}$$

Experimental and theoretical work shows clearly that higher

terms in B^2 must be considered with smaller samples (large buckling). This is due to transport theory corrections to the elementary diffusion theory approximation, and the "diffusion cooling" of neutrons.

Nelkin (48) has given an exact theoretical treatment to the above mentioned effect. His analysis of the decay of a thermalized neutron pulse was based on the assumption of an infinite medium in which the neutrons have reached the fundamental mode. The transport equation was solved by expanding the decay constant and neutron spectrum in a power series in the Fourier-transform variable. In order to obtain an expression for the decay constant to order B^4 term, he further assumed an isotropic scattering in a $\frac{1}{v}$ absorber. The coefficient C of B^4 contains two components:

$$C = C_D + C_t$$

C_D describes the "diffusion cooling" of the spectrum and C_t the "transport theory" contribution to the B^4 term.

The coefficient of B^2 in the expression:

$$\lambda = \lambda_o + D_o B^2 - CB^4 + \ldots$$

is the diffusion coefficient as usually defined. For a velocity dependent cross section this involves the Maxwellian average of the transport mean free path

$$\lambda_{tr} = \frac{A}{L\rho} \frac{\int \frac{E\, e^{-E/kT}}{\sigma_{tr}(E,T)}\, dE}{\int E\, e^{-E/kT} dE}$$

where

L = Avogadro's number,

A = Atomic weight of the moderator,

ρ = Density of the Moderator,

$\sigma_{tr}(E,T)$ = Microscopic transport cross section for neutrons of energy E in moderator at temperature T°K

k = Boltzmann's constant

The analysis of λ vs B^2 curves measured for moderators yields the absorption cross section, the average thermal neutron diffusion coefficient, and the diffusion cooling constant.

IV. EXPERIMENTAL ARRANGEMENT

A. The Pulsed Neutron Source

The pulsed neutron source consisted of a 250-kv Cockcroft-Walton accelerator using the $_1H^2(d,n)_2He^3$ reaction. Deuterium gas was supplied to a radio frequency ion source, operating at approximately 80 mc/sec. The oscillator and auxiliary equipment were built to ORTEC specifications. The deuteron beam was focused along a drift tube by an einzel lens and struck a target made of a water-cooled copper disk with a surface deposit of zirconium. The target was replenished by bombarding it occasionally with a steady beam of about 300 - 400μA for approximately 30 minutes. The D-D reaction, rather than the D-T reaction, was used to minimize the background due to photoneutrons from the heavy hydrogen in the mixtures of H_2O - D_2O. High energy gamma rays producing photoneutron reactions arise from the decay of $_7N^{16}$ which is produced from the $_8O^{16}(n,p)_7N^{16}$ reaction. The threshold energy for this reaction is about 10.5 Mev. By using the 2.5 Mev D-D neutrons this potential source of background was eliminated.

To pulse the neutron source a steady potential of about 700 volts was applied across a pair of deflecting plates situated between the einzel lens and the accelerating electrodes, to deflect the deuteron beam off the target. Pulses of the desired length and of opposite polarity were

then applied to cancel the deflecting voltage and permit the beam to enter the accelerating tube. Burst lengths of 10, 25, 80, and 170 microseconds and repetition rates from 12 to 1000 pulses per second were available. The pulsing system utilized a three centimeter wavelength microwave link to the high voltage terminal. Operation of the pulsing system and the einzel lens has been described previously (2).

Figure 1 is a diagram of the experimental arrangement and apparatus.

B. Moderator Assembly

In the liquid state the moderator materials were contained in aluminum cylindrical cans with dimensions such that the ratio of height to diameter was approximately equal to one.

Samples of moderator in the solid state (ice) were wrapped with aluminum foil to prevent sublimation of the ice. All moderator samples were covered with 30 mils of cadmium except for a small opening which allowed the detector to come in contact with the moderator.

The pulsed measurements were carried out in a large paraffin shield, Figure 2, which served both as a thermal insulator for the refrigeration chamber and an absorber of scattered neutrons. The cooling coils for the refrigerator were wound in a helix at the outside edge of the chamber. A small fan mounted in the top cover of the tank circulated

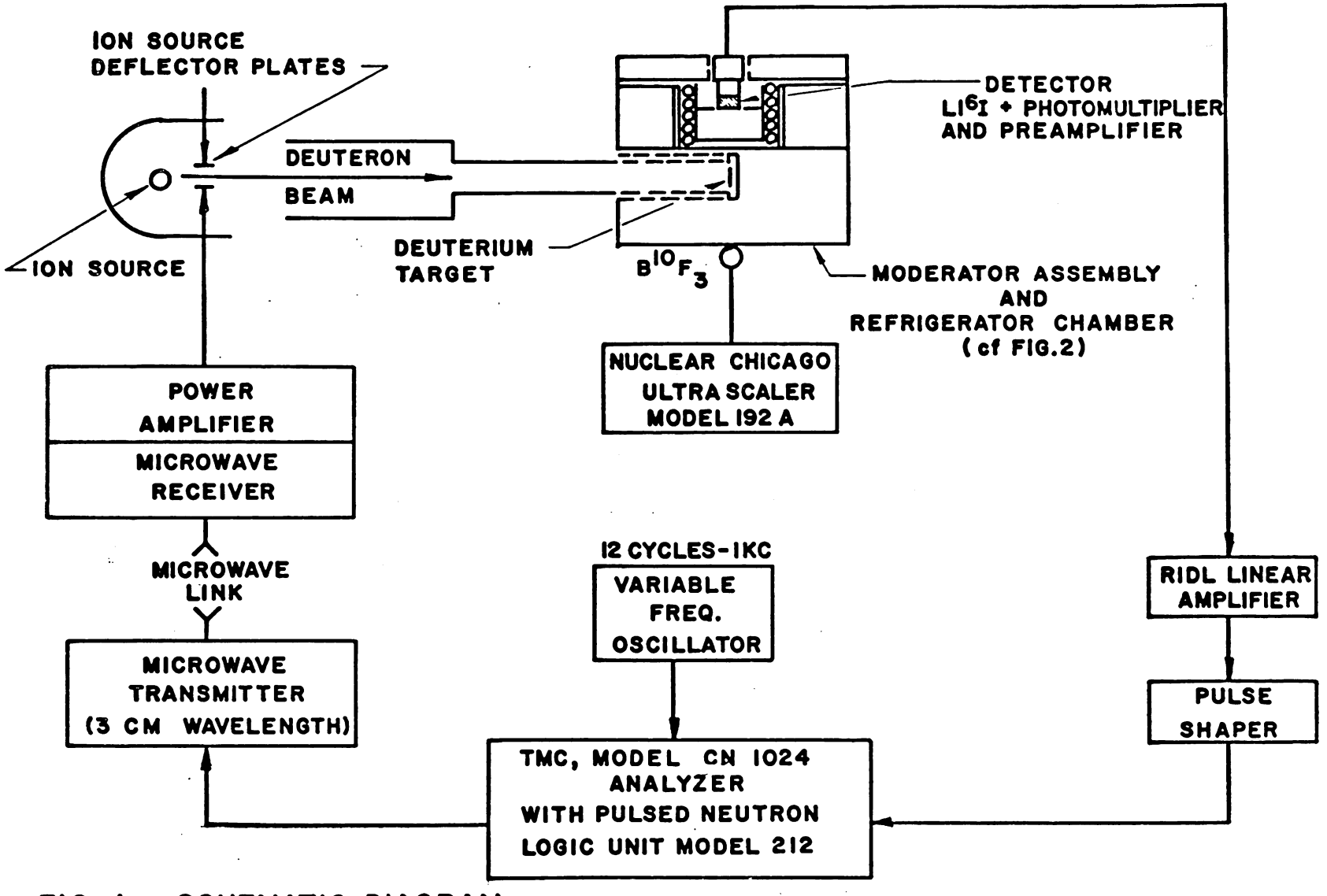

FIG. I. SCHEMATIC DIAGRAM
EXPERIMENTAL ARRANGEMENT

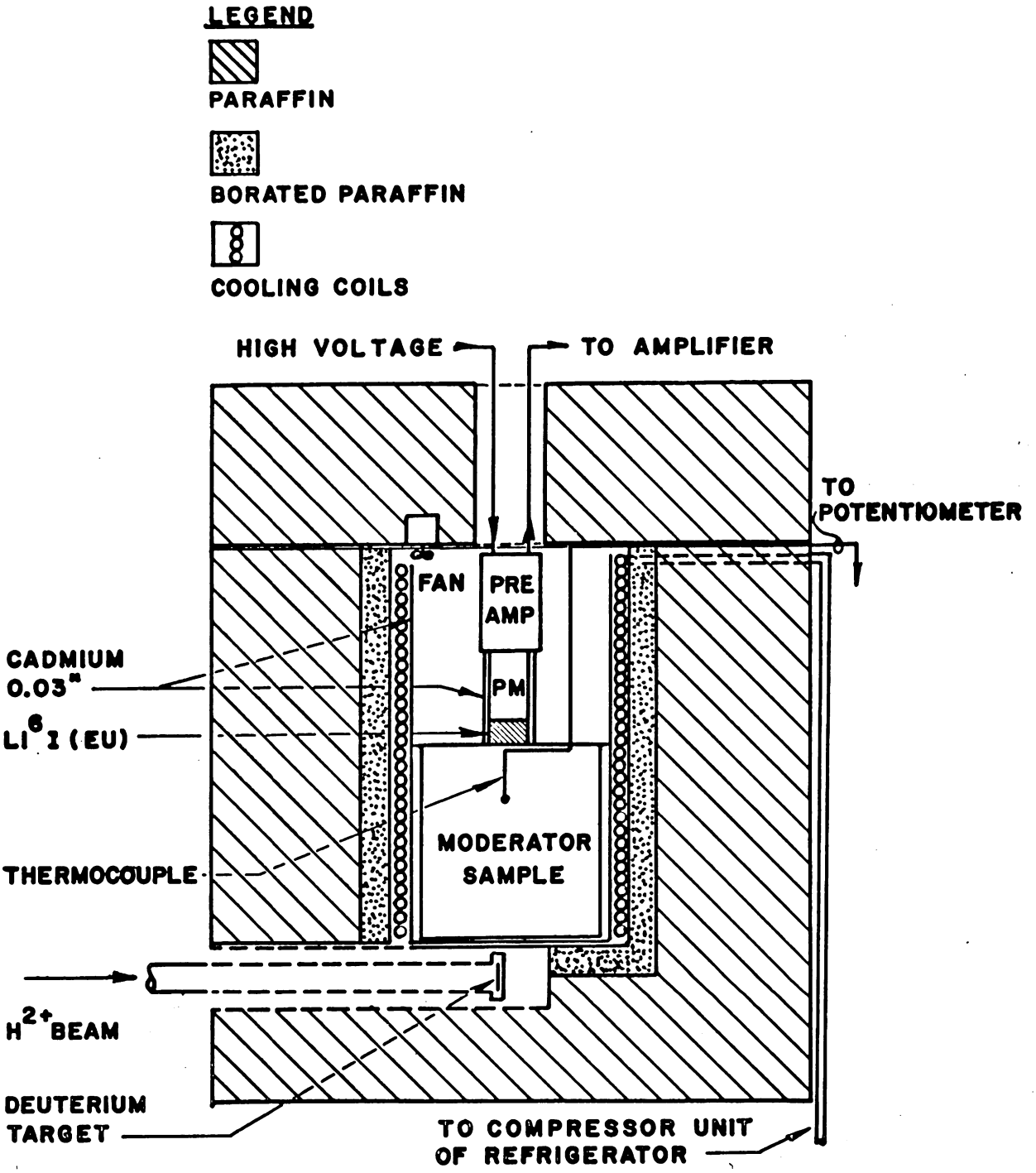

FIG. 2. SCHEMATIC DIAGRAM
MODERATOR ASSEMBLY & REFRIGERATOR CHAMBER

air which maintained a uniform temperature in the chamber and reduced frosting of the coils. The moderator temperature was measured with a copper-constantan thermocouple, one end of which was placed in the moderator and the other in an ice-water mixture. The thermocouples in the liquid and small ice samples were placed approximately at the center of the moderator and in the large ice samples at a depth of about three inches. This method of measuring temperature permitted determination of the moderator temperature to an accuracy better than $\pm$ 0.5°C. The presence of the thermocouple did not affect the values of the decay constant, λ.

C. Procedure for Making Uniform Ice Samples

Preliminary experiments with water were performed to determine methods for making ice free of entraped air and for adjusting the length of the frozen sample. For each ice cylinder an aluminum can one and one-half to two times the desired height was made. The liquid was transferred into the aluminum cylinder to a level about one and one-half times the desired height. The cylinder was placed in the vacuum system shown in Figure 3 and the system was pumped for several hours with a mechanical fore pump. The vacuum chamber and sample were then placed under vacuum in a deep freezer at -20°C. Once freezing was complete, usually after two days, the aluminum container was removed, covered,

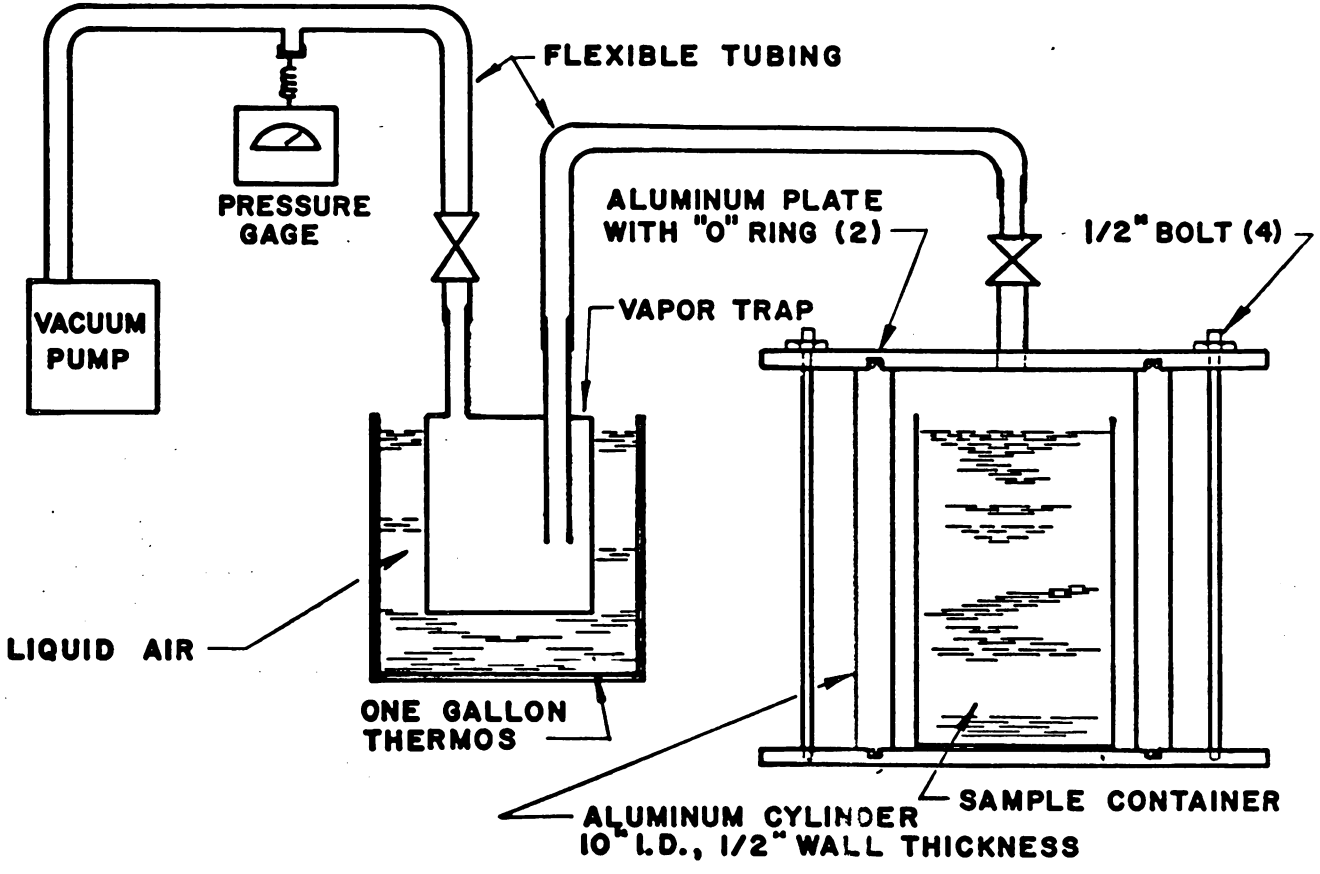

FIG. 3 SCHEMATIC DIAGRAM
SYSTEM USED FOR MAKING ICE CYLINDERS

and placed at room temperature until the ice was free to slide out of the can. Approximately the top one-third portion of the ice sample contained entraped air, but the remaining portion was clear ice. A sharp blow separated the two parts. The desired portion was left in the container and inverted in a flat tray placed over a heated plate to smooth the rough end. The ice cylinders thus formed were wrapped with aluminum foil, labelled, and placed in a deep freezer at -20°C.

Care was exercised to recover the D_2O vapor in the cold trap.

D. Neutron-Detection System

A small $Li^6I(Eu)$ crystal one inch diameter by two millimeter thick coupled to a Du Mont 6467 photomultiplier tube was used to measure thermal neutrons leaking from the sample. The photomultiplier and crystal assembly were wrapped with a cadmium sleeve except for the end which was in contact with the moderator. The detector assembly was brought to the desired temperature by gradual cooling. In order to prevent sudden exposure to room temperature during a sample change, an enclosure, maintained at the refrigerator temperature, was made in the top cover of the tank. Before the tank was opened the detector was retracted in this enclosure.

A $B^{10}F_3$ counter (Reuter-Stokes, Model RSN-7A) in

conjunction with an ultra-scaler (Nuclear Chicago, Model 192A) was used to monitor the neutron yield. Pulses from the detector assembly were fed through a preamplifier into a linear pulse amplifier and integral discriminator. Positive pulses from the discriminator were shaped and fed to the input of the time analyzer.

The time analyzer was a Technical Measurements Corporation Model 212 pulsed neutron logic unit used with a Model CN-1024 TMC Computer. The logic unit has a channel width variable from 10 to 2560 microseconds and also provides a background channel, a source trigger pulse after the background measurement, and a delay period during which time a neutron burst may be injected into the moderator sample. The background channel width and the delay period can be varied by 2, 4, 8, ..., 256 times the analysis channel width. At the end of the delay period the first analysis channel opens for counting. There is a constant 10 microseconds lapse time between channels to allow for storage in the memory of the computer unit. Pulses from a variable frequency pulse generator were used to trigger the analyzer. The data stored in the computer unit was read out on paper tape by a Hewlett Packard Digital printer.

Periodic checks were made on the pulse shape and resolution of the detector assembly. An additional check on the overall performance of the counting system was made by

using random pulses from a Pu-Be neutron source. Since these pulses are random in time a flat spectral response was obtained.

V. DATA ANALYSIS

Values of the decay constant, λ, and its standard deviation were determined from the measured neutron decay by use of a least squares fit computer program based on Peierl's statistical method (50). For data taken at large geometries (low buckling values) the decay constants were evaluated by dropping successive counting channels to verify that a fundamental mode was reached.

Figure 4 is a typical neutron density decay curve.

The infinite medium decay constants, λ_o, for H_2O, H_2O ice, and the H_2O - D_2O mixtures were obtained by a three-parameter least squares fit of the experimental data to the expression:

$$\lambda = \lambda_o + D_o B^2 - CB^4$$

Due to the limited number of data points for small buckling values, the value of λ_o for heavy water was calculated from the reported value and the known density. The values of λ_o for light-heavy water mixtures as found above were lower than the corresponding values computed from $\lambda_o(H_2O)$ and $\lambda_o(D_2O)$ in the same proportion as the mixtures. The range of discrepancy varied from about 60% for the 20% D_2O to 10% for the 80% D_2O mixtures, respectively.

The coefficients D_o and C and their standard deviation were obtained for each temperature by a least squares fit to the above expression after rearranging:

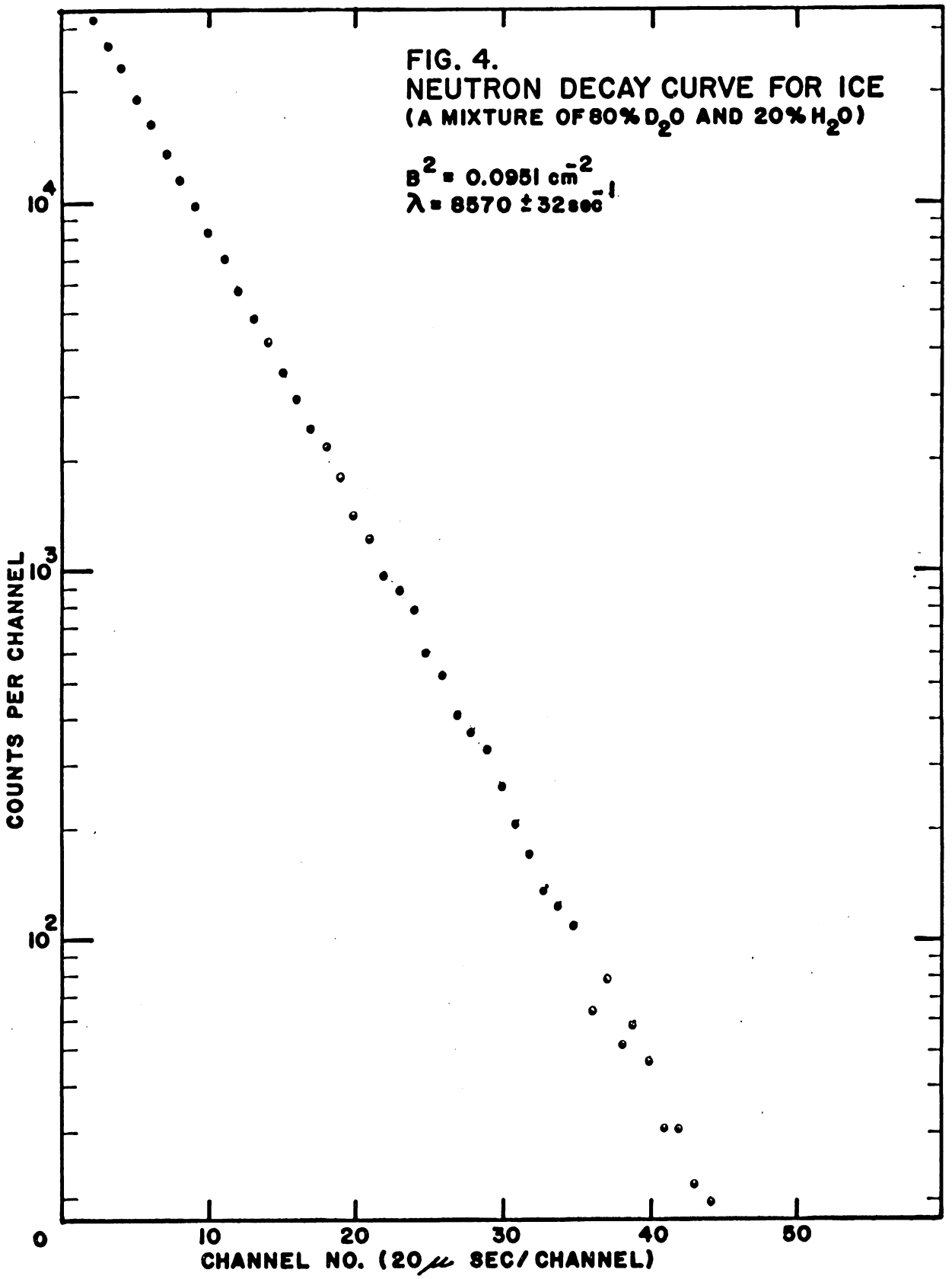
FIG. 4.
NEUTRON DECAY CURVE FOR ICE
(A MIXTURE OF 80% D_2O AND 20% H_2O)
$B^2 = 0.0951\ cm^{-2}$
$\lambda = 8570 \pm 32\ sec^{-1}$
COUNTS PER CHANNEL
10^4
10^3
10^2
0
10
20
30
40
50
CHANNEL NO. (20 μ SEC/ CHANNEL)

$$\lambda - \lambda_o = D_o B^2 - CB^4$$

Where B^2 is the buckling of the sample, D_o is the average diffusion coefficient, and C is the diffusion cooling constant. These calculations were performed by the computer using as input the experimental decay constants, the dimensions of the moderator sample, the value of λ_o and a trial value for the transport mean free path, λ_{tr}. An iterative procedure (15) was used in the computer program to make the value of D_o used in the expression for the extrapolation distance ($\epsilon = 2.131\ D_o/\bar{v}$) consistant with the results of the least squares fit.

For water and heavy water, the initial values of λ_{tr} pertaining to different temperatures were calculated by means of an empirical relation reported by Deutsch (51) based on the thermal diffusion length. The values of λ_{tr} for the H_2O - D_2O mixtures were calculated using the H^1, D^2, and O^{16} scattering cross sections and the relations:

$$\lambda = \frac{1}{\Sigma_{tr}}$$

where

$$\Sigma_{tr} = \Sigma_s(1 - \mu_o) = \sum_i N_i\ (\sigma_s)_i\ (1 - \mu_o)_i$$

Σ_s = macroscopic scattering cross section

μ_o = average value of the cosine of the scattering angle in the laboratory system

N_i = number of atoms per cm^3 of i^{th} component in the moderator

$(\sigma_s)_i$ = microscopic scattering cross section for i^{th} component in the moderator

The value for μ_o for hydrogen in water was calculated by the Radkowsky prescription (52). Use of the fundamental expression (53):

$$\lambda_{tr} = \frac{1}{\sum_i a_i / \text{tr},i}$$

where

$\lambda_{tr,i}$ = the transport mean free path of the i^{th} component of the mixture

α_i = the fractional volume of the i^{th} component of the mixture

gives the same results.

Both methods for calculating λ_{tr} yield results in good agreement with values obtained by the iterative procedure.

VI. RESULTS AND DISCUSSION

The measured decay constants, the dimensions of the samples, and iterated buckling values used in the least squares analysis to obtain the parameters λ_o, D_o, and C are given in Tables V - IX for the various concentrations of H_2O - D_2O mixtures. Table IV shows the parameters λ_c, D_o, and C. Also included in this table is the transport mean free path and the standard error of the above parameters.

Since oxygen has a very small absorption cross section and since the cross section for hydrogen varies as $\frac{1}{v}$, the value of the infinite medium decay constant, $\lambda_o = \Sigma_a v$, is independent of temperature. The results of the present investigations verified, within the limits of experimental error, the temperature independence of λ_o as is shown in Figures 5 - 9. The light water values of λ_o at 1°C in the liquid state and ice at -20°C are in good agreement with the value reported by Silver (12).

The value of D_o for H_2O ice at -20°C is in excellent agreement with the value calculated from Silver's data. D_o for H_2O at room temperature is in good agreement with published experimental results as shown in Table I.

The value of the diffusion cooling constant, C, for H_2O and H_2O ice is higher than the reported experimental results (8, 12).

In the case of D_2O at room temperature (21°C), the value

TABLE IV

Experimental Results - Thermal Neutron Diffusion Parameters

Percent D_2O	$D_o \times 10^{-4}$ ($cm^2 sec^{-1}$)	$C \times 10^{-5}$ ($cm^4 sec^{-1}$)	$\Sigma_a v$ (sec^{-1})	λ_{tr} (cm)	Temp (°C)
0	3.580 ± 0.066	0.0709 ± 0.0145	4462 ± 547	0.433	21
0	3.263 ± 0.075	0.0691 ± 0.0162	4462 ± 547	0.395	1
0	3.131 ± 0.069	0.0972 ± 0.0145	4325 ± 235	0.378	-20 (ice)
20	5.222 ± 0.119	0.2336 ± 0.0282	2186 ± 373	0.631	21
20	4.792 ± 0.041	0.2267 ± 0.0095	2186 ± 373	0.597	1
20	4.411 ± 0.084	0.1921 ± 0.0185	2006 ± 343	0.533	-20 (ice)
50	7.156 ± 0.180	0.4422 ± 0.0629	1036 ± 129	0.865	21
50	6.295 ± 0.154	0.3137 ± 0.0522	1036 ± 129	0.761	5
50	6.005 ± 0.053	0.3088 ± 0.0182	950 ± 119	0.726	-20 (ice)
80	11.458 ± 0.1151	1.3401 ± 0.0675	212 ± 110	1.385	21
80	10.720 ± 0.1788	1.3333 ± 0.1026	212 ± 110	1.296	5
80	10.149 ± 0.0619	1.2630 ± 0.0345	186 ± 97	1.227	-20 (ice)
100	19.662 ± 1.367	3.5551 ± 1.8574	19	2.681	21
100	19.630 ± 0.637	4.8859 ± 0.4612	19	2.373	5
100	20.534 ± 0.768	4.7230 ± 0.6526	17.2	2.472	-20 (ice)

of D_0 obtained for pure D_2O (2) is in good agreement with that reported by Daughtry and Waltner (21). Whereas the value of C is lower than the reported experimental values. The values of D_0 for pure D_2O at 5°C and D_2O ice at -20°C, after temperature and density corrections, lie between the experimental value of Kussmaul and Meister (17) and the theoretical value by Honeck and Michael (18). The value of D_0 by Baumann (20) is larger than the theoretical value by Honeck and Michael.

The values of C for D_2O at 5°C and D_2O ice at -20°C are lower than the value by Kussmaul and Meister (17) and higher than the experimental values by Daughtry et al. and the theoretical values by Honeck and Michael.

There is very little data available on the parameters of H_2O - D_2O mixtures (22) and no valid comparison can be made.

The curve of λ versus B^2 for D_2O ice, Figure 9, is higher than the corresponding curves at 5° and 21°C. This shift is primarily due to density difference between the liquid and solid states. Since in the expression

$$\lambda = \lambda_0 + D_0B^2 - CB^4$$

λ_0 is negligibly small, and since D_0 varies inversely with the moderator density, the second term on the right hand side is the dominant term. The value of D_0 for D_2O ice at -20°C is larger than the values at 5°C and 21°C, resulting

in the shift in the λ versus B^2 curve.

The standard errors listed in Table IV were due to contributions from the uncertainties in the decay constants, the bucklings, and the values of the infinite medium decay constants, λ_o. Neutron return from the tank and backscattering from the tank metal container contributed to the uncertainty in the decay constant.

The errors in the buckling values are from the following quantities:

1. The aluminum cans were not perfect right circular cylinders. The dimensions given are average values over several measurements of the radii and heights.
2. The density of the solid samples; some of the ice samples contained a small conic section of entrapped air.
3. The extrapolation distance, ε, where

   $$\varepsilon = 0.7104\lambda_{tr} = 2.131\ D_o/\bar{v}$$

   is proportional to the error in D_o. The errors in B^2 due to uncertainties in ε are significant for large buckling values (small sizes).

An additional error may be attributed to the temperature of the samples. Although, temperature equilibrium was established for large samples before a measurement was made, it was difficult to control the temperature variation of the small samples to better than $\pm$ 2°C.

Figure 10 shows D_o versus heavy water concentration as a function of temperature.

TABLE V

Measured Decay Constants for H_2O

r(cm)	H(cm)	B^2 (cm^{-2})	λ (sec^{-1})	Temp. (°C)
11.69	24.57	0.0558	6358 ± 48	
9.41	19.22	0.0863	7212 ± 48	
8.07	16.07	0.1178	8515 ± 27	
6.38	12.70	0.1852	10890 ± 72	21
5.36	10.75	0.2563	13313 ± 44	
4.24	8.61	0.3958	17576 ± 114	
3.56	7.14	0.5515	21995 ± 66	
11.69	24.57	0.0560	6103 ± 31	
9.41	19.22	0.0868	7211 ± 48	
8.07	16.07	0.1186	7956 ± 51	
6.38	12.70	0.1867	10369 ± 76	1.0
5.36	10.75	0.2588	12665 ± 194	
4.24	8.61	0.4006	16414 ± 115	
3.56	7.14	0.5594	20518 ± 116	
11.69	20.48	0.0628	6336 ± 32	
8.85	15.72	0.1069	7421 ± 37	
7.71	15.88	0.1276	8190 ± 39	
6.34	12.89	0.1870	9710 ± 50	-20
5.31	10.52	0.2667	12251 ± 74	ice
4.16	8.33	0.4202	15607 ± 88	
3.50	7.18	0.5734	19119 ± 124	

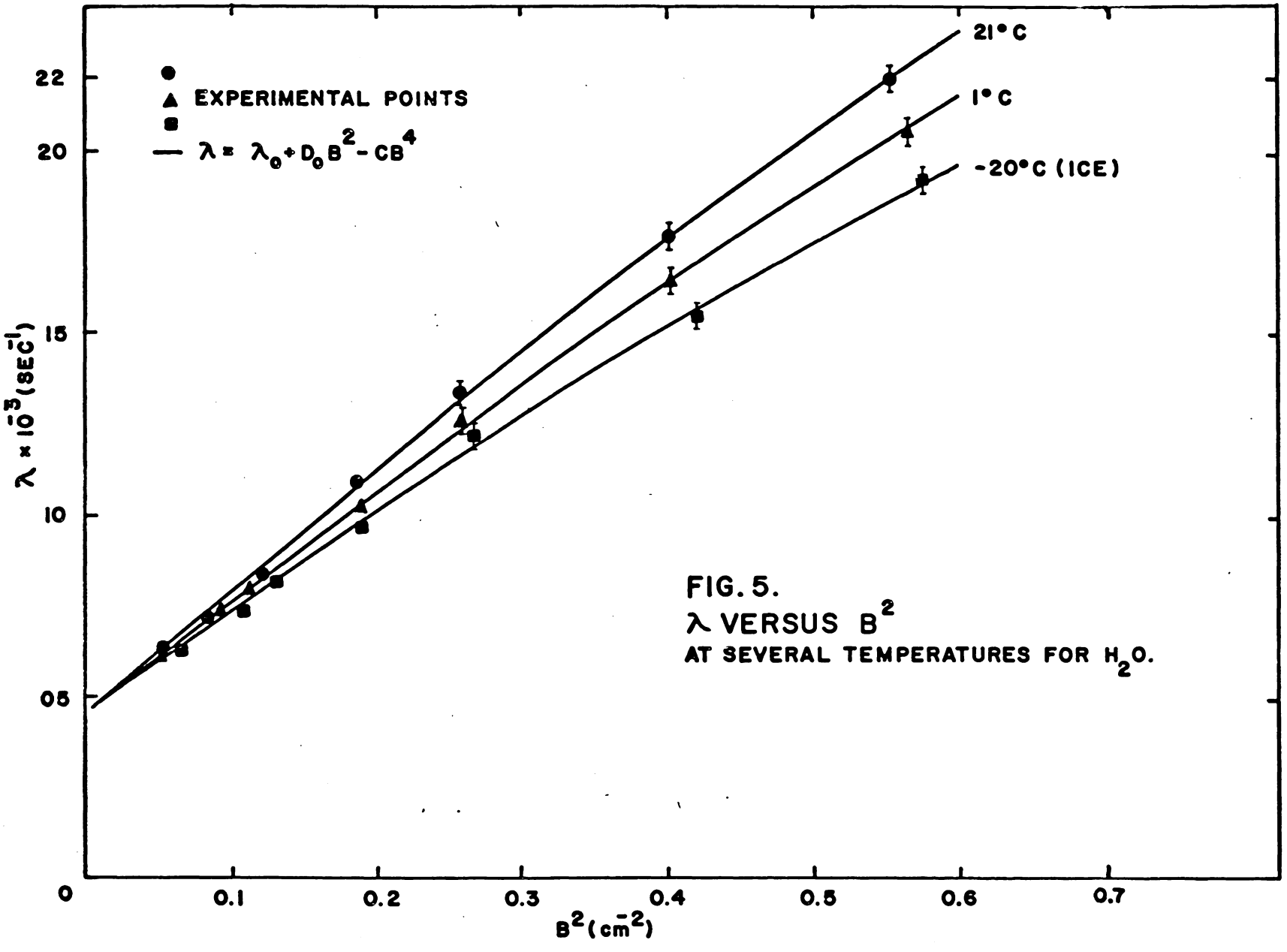

FIG. 5.
λ VERSUS B^2
AT SEVERAL TEMPERATURES FOR H_2O.

TABLE VI

Measured Decay Constants for 20 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\lambda(sec^{-1})$	Temp. (°C)
11.69	24.57	0.0545	4840 ± 23	
9.41	19.22	0.0839	6478 ± 34	|
8.07	16.07	0.1139	7477 ± 47	|
6.38	12.70	0.1776	10181 ± 62	21
5.36	10.75	0.2440	13407 ± 84	|
4.24	8.61	0.3725	18521 ± 99	|
3.56	7.14	0.5135	22538 ± 122	
11.69	24.57	0.0548	4568 ± 24	|
9.41	19.22	0.0845	5927 ± 32	|
8.07	16.07	0.1149	7039 ± 36	
6.38	12.70	0.1796	9860 ± 25	1.0
5.36	10.75	0.2472	12558 ± 92	|
4.24	8.61	0.3784	16922 ± 156	|
3.56	7.14	0.5231	20841 ± 66	
11.69	24.01	0.0558	4259 ± 23	|
8.85	16.43	0.1013	6333 ± 33	|
7.71	15.35	0.1266	7231 ± 27	
6.34	12.39	0.1852	9543 ± 36	-20
5.31	10.45	0.2574	12373 ± 54	ice
4.16	9.55	0.3736	15913 ± 45	
3.50	6.99	0.5497	20520 ± 57	|

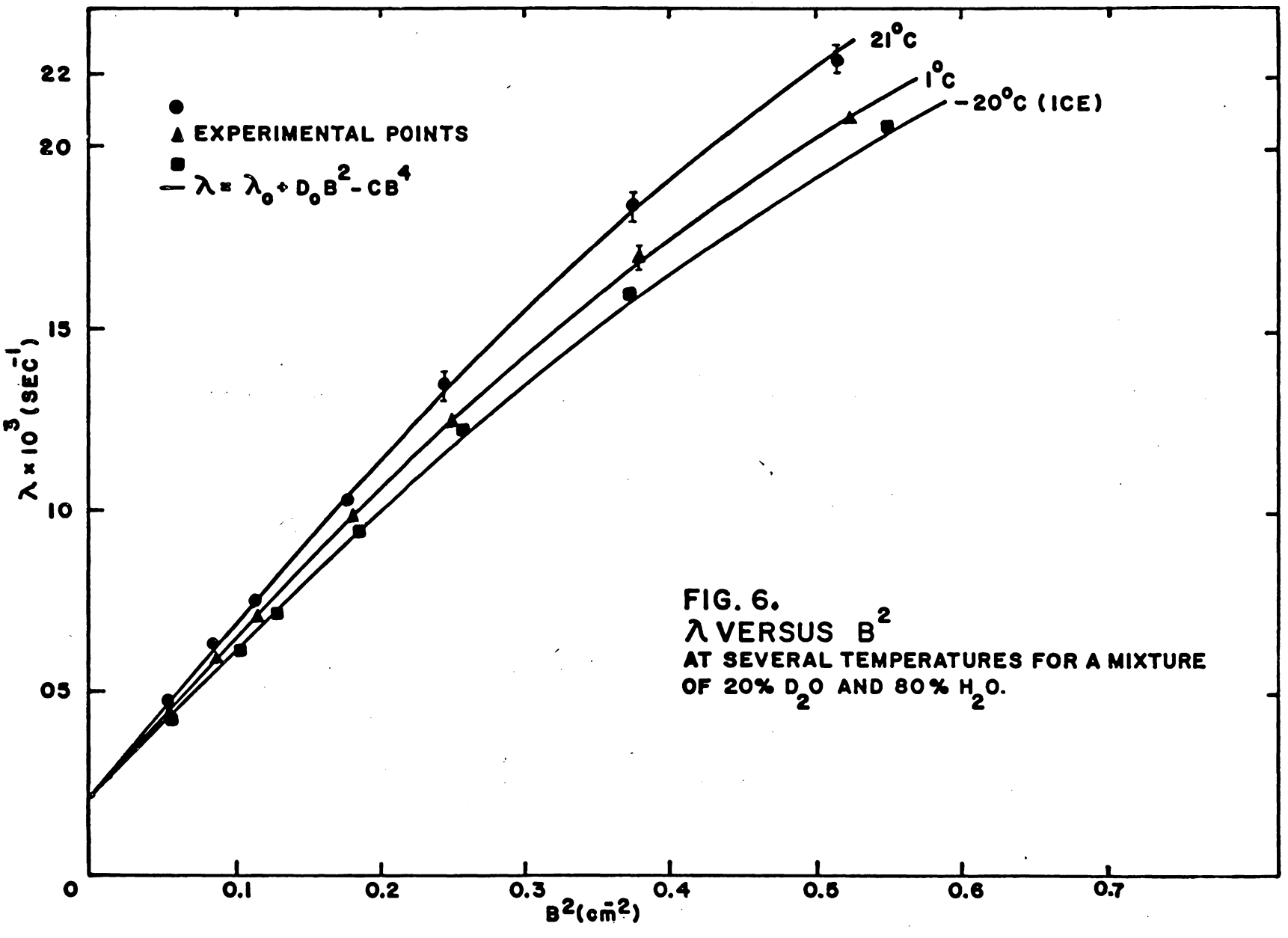

FIG. 6.
λ VERSUS B^2
AT SEVERAL TEMPERATURES FOR A MIXTURE OF 20% D_2O AND 80% H_2O.

TABLE VII

Measured Decay Constants for 50 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\lambda(sec^{-1})$	Temp. (°C)
11.69	24.57	0.0530	4419 ± 32	21
9.41	19.22	0.0812	6491 ± 25	
8.07	16.07	0.1096	7916 ± 42	
6.38	12.70	0.1693	12131 ± 58	
5.36	10.75	0.2305	15162 ± 110	
4.24	8.61	0.3476	20413 ± 113	
11.69	15.66	0.0739	5096 ± 31	5.0
9.41	19.22	0.0824	5750 ± 28	
8.07	16.07	0.1115	7555 ± 45	
6.38	12.70	0.1729	11055 ± 51	
5.36	10.75	0.2365	14306 ± 78	
4.24	8.61	0.3584	19370 ± 104	
11.69	17.70	0.0670	4872 ± 36	-20 ice
8.85	15.64	0.1015	6807 ± 36	
7.71	14.61	0.1260	7984 ± 33	
6.34	11.12	0.1898	11358 ± 44	
5.31	11.07	0.2379	13389 ± 33	
4.16	9.05	0.3617	18645 ± 54	

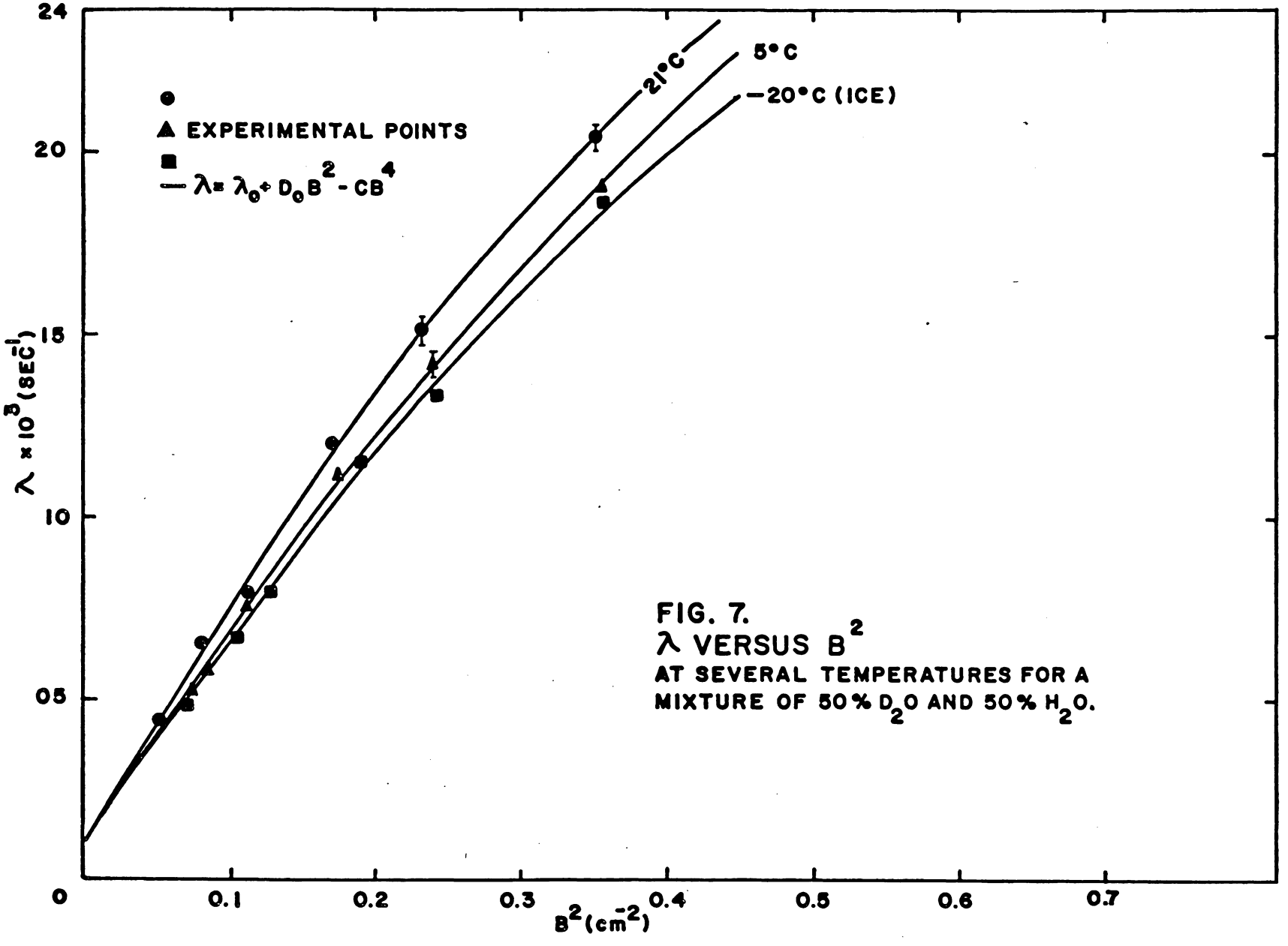

FIG. 7.
λ VERSUS B^2
AT SEVERAL TEMPERATURES FOR A MIXTURE OF 50% D_2O AND 50% H_2O.

TABLE VIII

Measured Decay Constants for 80 Percent D_2O

r(cm)	H(cm)	$B^2(cm^{-2})$	$\lambda(sec^{-1})$	Temp. (°C)
11.69	22.38	0.0527	5705 ± 37	|
9.41	19.22	0.0755	8109 ± 48	
8.07	16.07	0.1009	10435 ± 52	21
6.38	12.70	0.1527	14586 ± 51	
5.36	10.75	0.2047	17998 ± 61	|
11.69	24.57	0.0505	5177 ± 28	|
9.41	19.22	0.0764	7476 ± 28	
8.07	16.07	0.1023	9795 ± 38	5.0
6.38	12.70	0.1554	13803 ± 39	
5.36	10.75	0.2088	16666 ± 70	|
11.69	20.08	0.0574	5572 ± 32	|
8.85	15.88	0.0930	8570 ± 32	
7.71	15.08	0.1135	10015 ± 47	-20 ice
6.34	12.94	0.1569	13054 ± 133	
5.31	10.40	0.2184	16310 ± 144	|

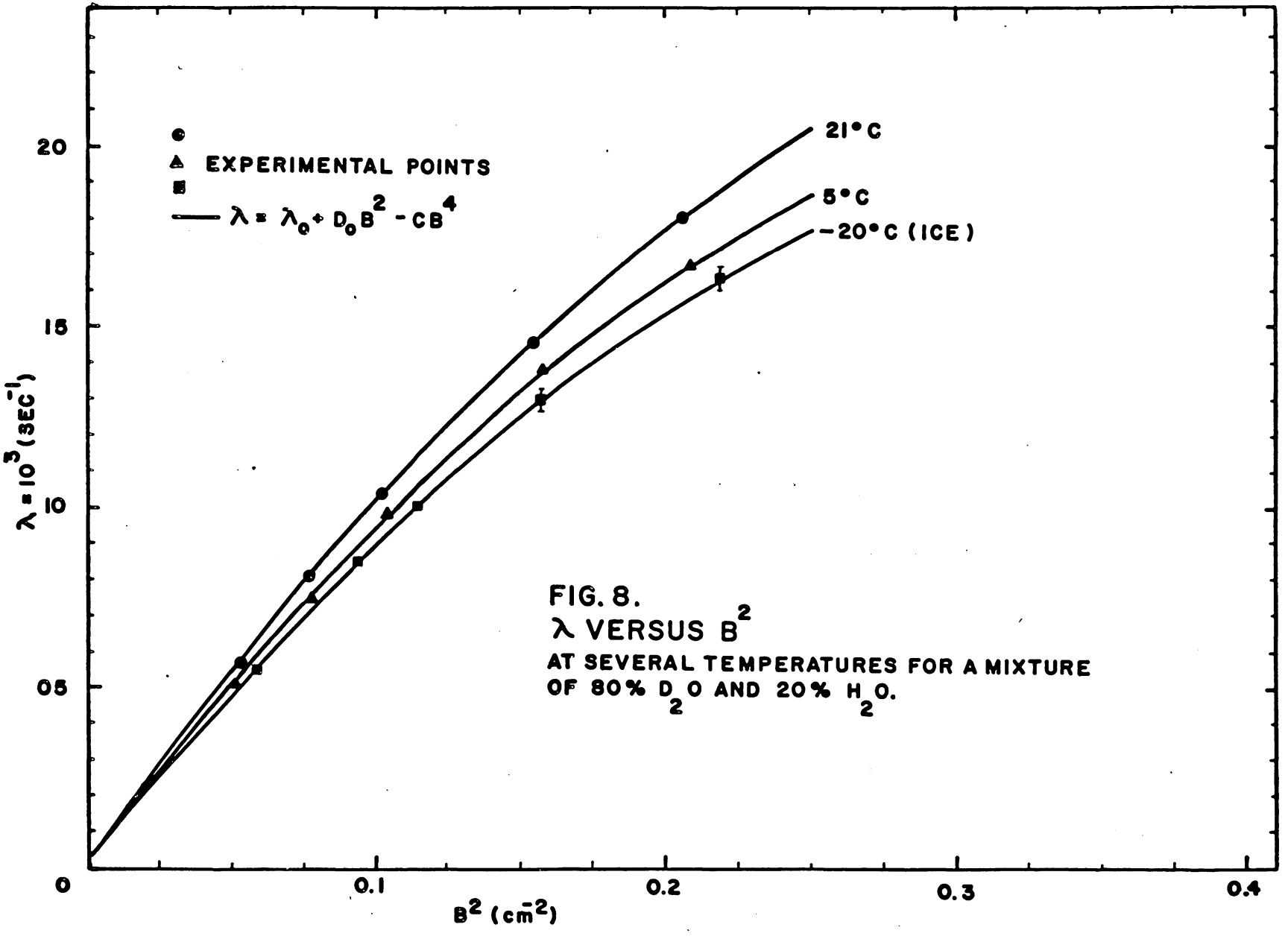

FIG. 8.
λ VERSUS B^2
AT SEVERAL TEMPERATURES FOR A MIXTURE OF 80% D_2O AND 20% H_2O.

TABLE IX

Measured Decay Constants for D_2O

r(cm)	H(cm)	B^2 (cm^{-2})	λ (sec^{-1})	Temp. (°C)
11.69	24.57	0.0453	8161 ± 100	|
11.69	16.83	0.0545	9917 ± 105	|
9.41	19.22	0.0638	10639 ± 111	21
8.83	16.39	0.0745	12394 ± 141	|
8.83	12.29	0.0884	14870 ± 151	|
11.69	24.57	0.0450	7442 ± 31	|
9.41	19.22	0.0663	10800 ± 127	
8.07	16.07	0.0869	13935 ± 136	5.0
6.38	12.70	0.1273	16901 ± 428	
5.36	10.75	0.1660	19144 ± 407	|
11.69	19.69	0.0503	8934 ± 37	|
8.85	15.88	0.0776	13654 ± 121	
7.71	12.54	0.1028	16659 ± 137	-20
6.34	12.38	0.1271	19091 ± 228	ice
4.16	9.45	0.2144	20105 ± 611	|

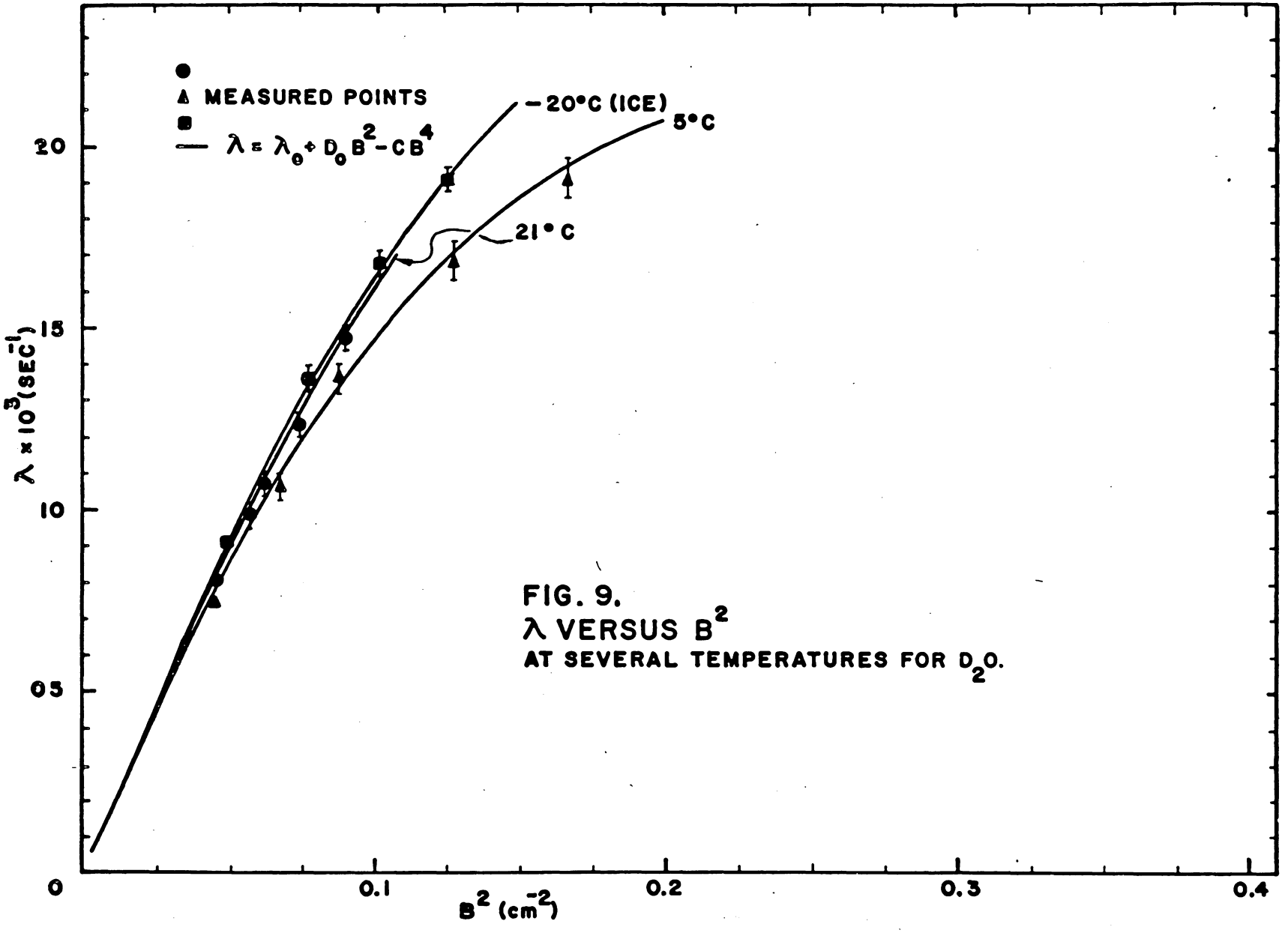

FIG. 9.
λ VERSUS B^2
AT SEVERAL TEMPERATURES FOR D_2O.

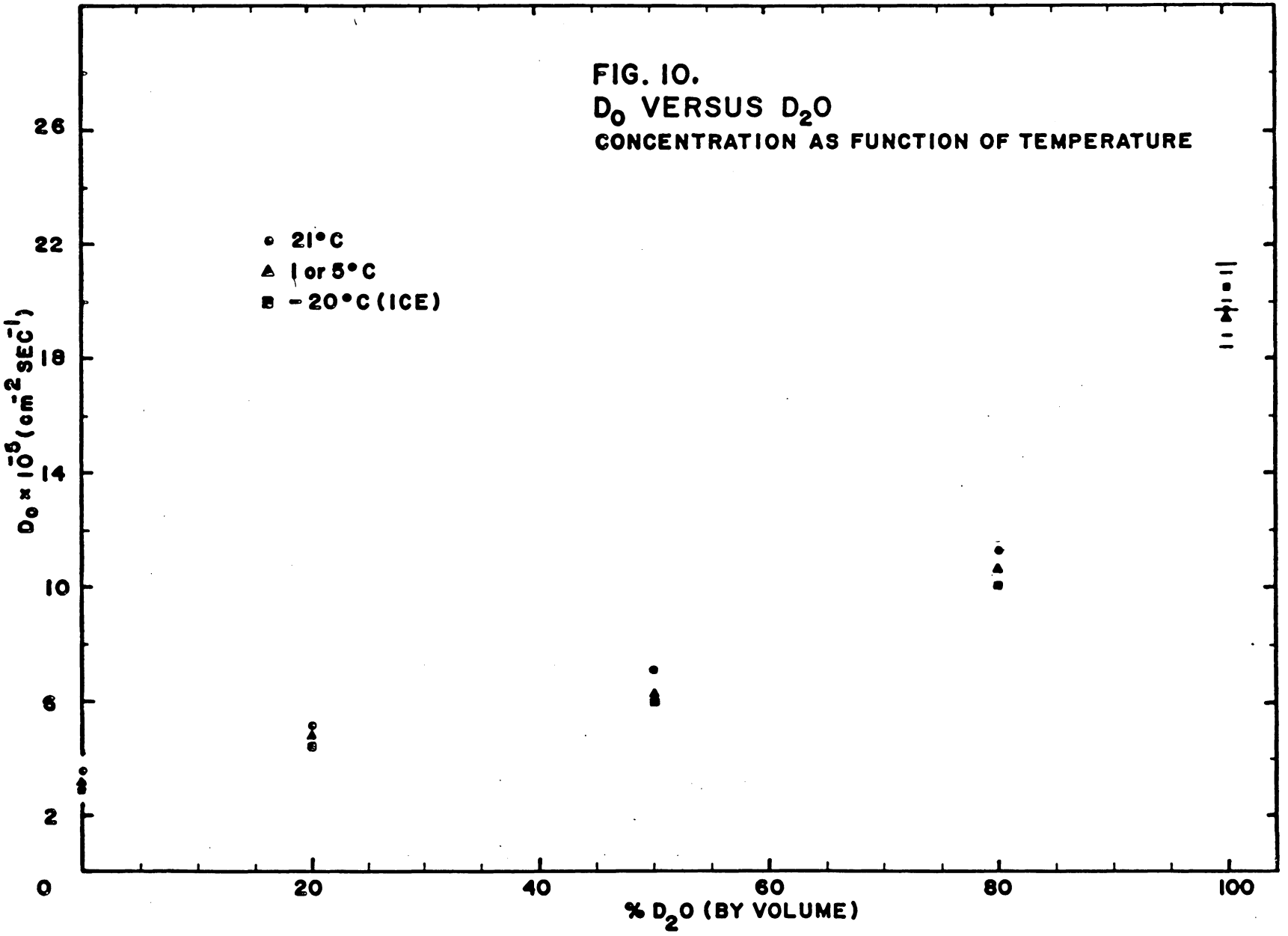
FIG. 10.
D_0 VERSUS D_2O
CONCENTRATION AS FUNCTION OF TEMPERATURE
21°C
1 or 5°C
-20°C (ICE)
$D_0 \times 10^{-5}$ (cm^{-2} sec^{-1})
0
2
6
10
14
18
22
26
0
20
40
60
80
100
% D_2O (BY VOLUME)

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X. APPENDIX

Determination of the D_2O Content of H_2O - D_2O Mixtures

The mole percent concentration of D_2O in each mixture was determined to an accuracy of $\pm$ 0.2% using an Abbe refractometer. Figure 11 is a graph of index of refraction versus percentage of D_2O. Calibration of the refractometer was made using the handbook index of refraction values for pure H_2O and D_2O and gravimetrically analyzed samples of D_2O - H_2O mixtures furnished by the Babcock and Wilcox Company. Table X lists the measured index of refraction values.

TABLE X

Index of Refraction for D_2O - H_2O Mixtures

D_2O Concentration Mole Percent	Index of Refraction
49.76 ± 0.02*	1.3303
60.40 ± 0.02*	1.32967
69.70 ± 0.05*	1.32925
85.50 ± 0.05*	1.32862
0†	1.33299
20	1.33156
50	1.33028
80	1.32896
100†	1.3281

* Samples furnished by the Babcock and Wilcox Company

† Handbook value at 20°C

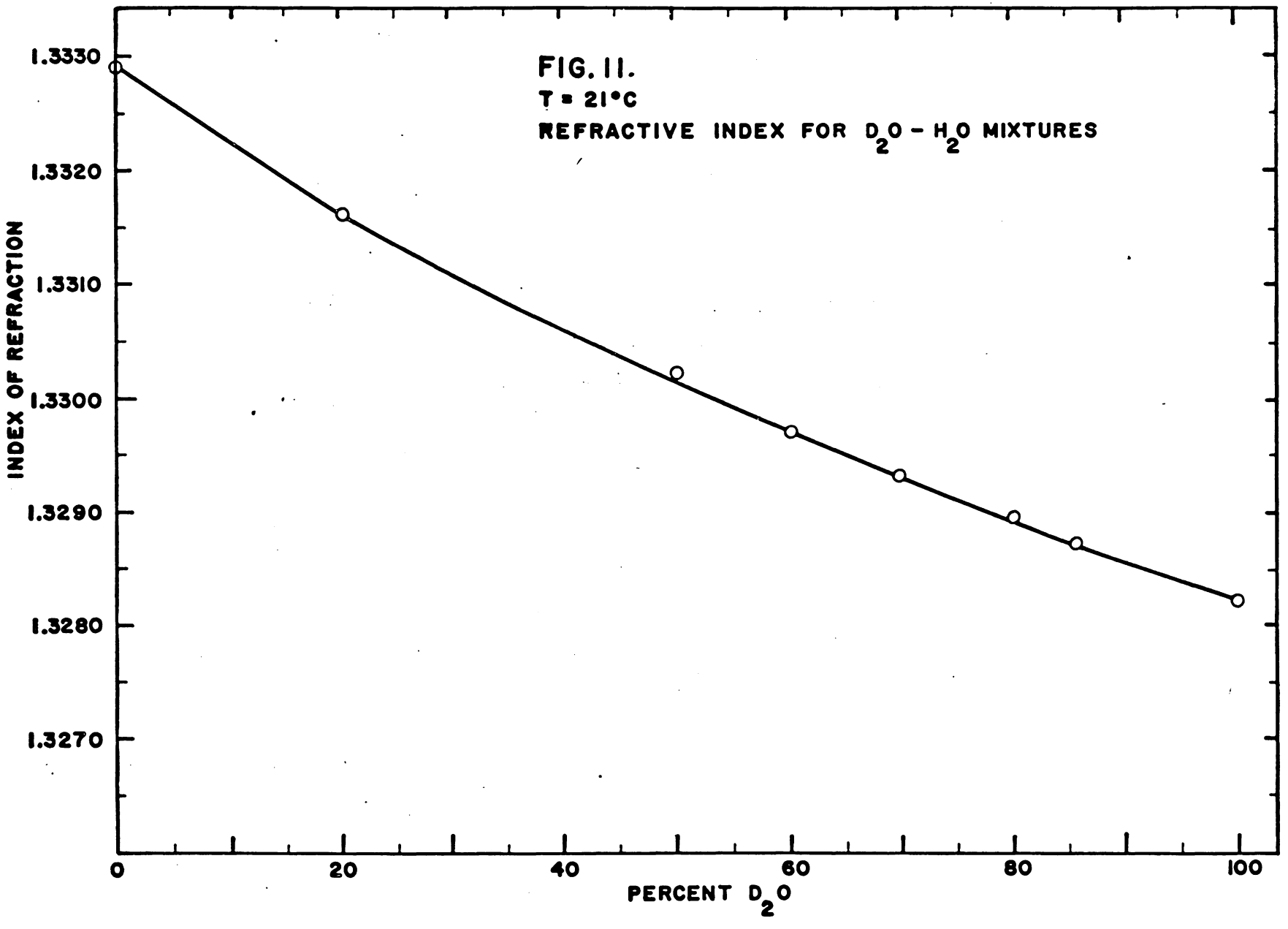
FIG. 11.
T = 21°C
REFRACTIVE INDEX FOR $D_2O - H_2O$ MIXTURES
INDEX OF REFRACTION
1.3330
1.3320
1.3310
1.3300
1.3290
1.3280
1.3270
0
20
40
60
80
100
PERCENT D_2O

ABSTRACT

Thermal neutron diffusion parameters of 0, 20, 50, 80 and 100 percent D_2O in mixtures of light and heavy water were measured by the pulsed neutron technique at room temperature (21°C), near the freezing point of the mixture in the liquid state, and at -20°C in the solid state (ice). A 250-kv pulsed Cockcroft-Walton accelerator, using neutrons from the $_1H^2(d,n)_2He^3$ reaction, was used to generate fast neutrons bursts.

The values of the infinite medium decay constant, λ_o, were computed by a least squares fit of λ versus B^2 data to the expression:

$$\lambda = \lambda + D_o B^2 - CB^4$$

The parameters D_o, C and λ_{tr} were determined from the above expression after rearranging:

$$\lambda - \lambda_o = D\ B^2 - CB^4$$

The resulting values of D_o, C, λ, and λ_{tr} at several temperatures are tabulated.