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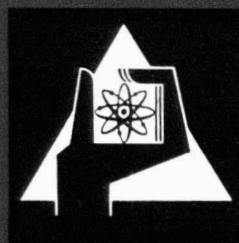
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I. Introduction

The combined pulsed source chopper technique has proved to be a valuable tool for the investigation of neutron spectra in moderators. As is well known, the time asymptotic equilibrium state in an infinite moderator with $\frac{1}{v}$-absorption is completely governed by the principle of detailed balance, which causes the spectrum to be a Maxwellian. To get information about the scattering properties of the moderator, it is therefore necessary to study deviations from this state by enforcing a distortion of the spectrum. This situation is realized when neutron leakage from the moderator is present. For small moderator assemblies the spectrum inside is distorted by diffusion cooling. A direct measurement of this phenomenon has been made by Beckurts [1] . For large moderators the leakage causes a distortion of the spectrum only near the surface. The leakage spectra correspond to those of the Milne problem without absorption and a Maxwellian source at infinity. This problem has been investigated experimentally and theoretically by various authors [1, 2, 3, 4, 5]. Of interest are the angular dependence of the spectra and how they are influenced by the detailed form of the scattering law.

A more direct method is to look into the approach to the equilibrium state, where the deviations are larger and therefore the influence of the scattering properties of the moderator becomes more apparent. Measurements of the time-dependence of neutron spectra have first been made by Barnard et al. [6] . For large moderators such investigations yield information mainly about the isotropic part of the scattering law, whereas diffusion cooled and leakage spectra are influenced by both transport effects and thermalization effects which causes a very complex dependence on the scattering law.

In this paper two different investigations of thermal neutron spectra by the combined pulsed source chopper technique are described.

The first part deals with time asymptotic leakage spectra. For a large moderator angular dependent measurements were made for water and leakage spectra perpendicular to the surface were measured for water, benzene, and Dowtherm A. Furthermore for water and benzene measurements of the perpendicular leakage spectra were performed for small slabs, where the spectra in the middle no longer are Maxwellian due to diffusion cooling.

The second part concerns investigations of the neutron moderation through measurements of the time behaviour of neutron spectra after the injection of a short pulse of fast neutrons into a moderator. The moderators investigated were zirconium hydride at room temperature and ice at liquid nitrogen and liquid hydrogen temperature.

II. Leakage spectra (J.K.)

1. Experimental methods

The pulsed neutron source and flight path have been described in [7]. The source, a 400 kev cascade deuteron accelerator, was developed by W.Eyrich [8]. At the end of the three meter flight path is the neutron detector bank of eight high pressure BF_3 counters. A cadmium chopper with a rotation rate of 7000 - 12000 RPM is used. An electronic pulse given from the chopper is delayed, and then used to trigger the accelerator and the time analyzer connected to the detector. Thus, by adjusting the time at which the accelerator pulse occurs, it is insured that the neutrons in the moderator are thermalized before the chopper opens. Three long counters were used as monitors, to determine the intensity of the source.

The measurements of the leakage spectra for neutrons emitted at angles from 0° to 72° from the surface normal were made on two large water samples, 15 cm x 30 cm x 30 cm and 12 cm x 22 cm x 22 cm. Fig.1 shows schematically the experimental set-up for this measurement. The different angles were obtained by turning the sample about the axis through the center of the area from which the neutrons were extracted. For each angular adjustment, the apparatus was moved so that the position of the target relative to the moderator sample remained constant. The area from which the neutrons were extracted was fixed by windows of various sizes, such that the area of the surface seen by the detector bank was identical for all angles.

In order to measure at larger angles from the normal to the surface, rather large distances (up to 14 cm) were necessary. This causes a considerable correction for the different flight times between the moderator surface and the chopper. To check the accuracy of this correction, measurements of the spectrum for leakage perpendicular to the surface were made at three distances. The spectra obtained from these measurements were identical.

The measurements of the spectra for neutrons emitted parallel to the surface were made on a 22 cm x 22 cm x 12 cm H_2O sample, using the assembly shown in fig.2. The extraction channel was two cm wide, and had a cadmium lining except for the last 8 mm from the extraction point; this lining insured that only neutrons from the extraction point could pass through the chopper. The water surface was sharply defined at the extraction point by an aluminium cross piece in contact with the surface. Measurements were made for channel intrusion depths below the surface of 2 and 4 mm.

2. Results

a) Perpendicular leakage spectra

The results for the leakage spectra perpendicular to the surface for H_2O, benzene, and Dowtherm A at room temperature are shown in fig.3 and 4. In these figures, and in fig.5, the solid curves are from theoretical calculations, which will be discussed in section 3.

The spectra for the large sample (30 cm x 30 cm x 15 cm) are compared in figures 3-IV and 3-V for the three moderators. These spectra are practically identical, although the frequency distributions of the three moderators are quite different [9, 10]. All three spectra are about 15 – 17% hotter than a Maxwellian distribution at moderator temperature, shown by the dashed curve in fig.3-I.

The effect of diffusion cooling, first observed directly in spectra measurements of Beckurts [1], is quite apparent in the spectra for small slabs. It can be seen in fig.4 that the maximum of the spectra for the smallest slabs is at about 0.025 ev, while that of the infinite medium is at 0.030 ev.

b) Angular dependent spectra

The results of the angular dependent spectra measurements are shown in fig.5, for various values of μ, the cosine of the angle θ between the emerging direction and the normal to the surface. The measurements for μ = 1.0, 0.7, 0.5, and 0.3 (θ = 0°, 45°, 60°, and 72°) are those from the 22 x 22 x 12 cm sample; the results with the 30 x 30 x 15 sample were essentially the same. There is very little angular dependence down to μ = 0.3, the spectra being all of nearly the same form. There is a very slight amount of cooling as μ decreases, which can be detected by dividing the μ = 1.0 curve into the other curves. As an example, fig.6 shows the quotient $\dfrac{\emptyset(\mu = 0.3, \ E)}{\emptyset(\mu = 1.0, \ E)}$. While the points scatter considerably, a definite trend toward higher values for lower energies can be seen, which indicates that the μ = 0.3 spectrum is cooler. For comparison, the quotient of two Maxwellians with T_0 differing by 2% is shown.

For μ = 0.0, the container described in the previous section for measuring spectra emerging parallel to surface was used. The spectra shown were obtained from measurements in which the extraction channel penetration into the surface was approximately 2 and 4 mm. The results for both intrusion depths are practically the same. The intensity for the 2 mm measurement was very low, therefore the points scatter considerably. Theoretical calculations [4] predict that the transient flux, which describes the variation in the spectra form near the surface, dies off quite rapidly for increasing distance from the surface. Furthermore, the disturbing effect of the extraction channel becomes greater for increasing penetration depth. Hence, the results must be interpreted with care. However, since the variation between the 4 and 2 mm measurements is minimal, it was felt justified to extrapolate, and presume that the 2 mm measurement gives essentially the form of the spectra for zero penetration, which is the case of interest. It can be seen that the spectra at the grazing angle are cooler than that for μ = 1.0. The dotted curve in fig.5-VI is a Maxwellian at moderator temperature; the spectrum is obviously hotter than this Maxwellian.

3. Theoretical calculations and comparison with measurements

The calculations for the buckling dependent perpendicular leakage spectra were done with ADALS, a program developed by R.Kladnik for the time asymptotic, angular dependent leakage spectra for infinite slabs. The theory for the program [11, 12] applies a variational technique. The influence of the surface on the spectra is described by a variational trial function, improved by one iteration.

The curves shown in figures 3 and 4 were calculated with ADALS. The water calculations shown are for the Goldman and Federighi version of the Nelkin kernel [13], while the Dowtherm A and benzene calculations were done with a kernel calculated by a Karlsruhe version of Pixse [14] from Gläser's scattering laws [9]. For water, calculations were also done with the Haywood kernel [10]; the results were essentially the same as with the Nelkin kernel. For the large sample, the calculated spectra for the organic moderators are slightly cooler than those for H_2O, but otherwise of practically the same form. As can be seen, the agreement between measurements and calculations is good for H_2O, while for benzene and Dowtherm A the calculated spectra are a little cooler than the measurements. For thin slabs, the agreement is satisfactory for all cases; thus ADALS describes quite well the influence of diffusion cooling. In general, it can be said that the agreement between ADALS calculations and experiment is satisfactory for the perpendicular leakage spectra.

The angular dependence of the leakage spectra calculated by ADALS, which predicts a Maxwellian for $\mu = 0.0$, was stronger than that which we detected experimentally. To obtain better agreement, modifications were made on another program developed by Kladnik, which treats the Milne problem for a semi-infinite medium with zero absorption and a Maxwellian source at infinity. The spectra from our angular measurements, made in a large geometry, correspond to this situation. The theory for this problem is described in [2]. Since the geometry effects for this case are much simpler, the trial function describing the influence of the surface can be more involved. For the trial function used in [2], with one iteration, the spectrum for $\mu = 0.0$ is a Maxwellian.

This program was modified by using a more complex trial function, and performing two iterations. For the perpendicular leakage spectra ($u = 1.0$), the results of the modified program agree well with the ADALS thick slab calculations. The results of a Haywood kernel calculation with the modified program are shown in fig.5. The theoretical spectra are 17% and 11% hotter than a Maxwellian for $u = 1.0$ and 0.0, respectively. As can be seen in the figure, the agreement with the measured spectra is satisfactory in all cases.

4. Summary

Measurements of the leakage spectra perpendicular to the surface of H_2O, benzene, and Dowtherm A for a large geometry show that for these moderators the influence of differences in the scattering law on the leakage spectra is very small. Whereas the frequency distributions of the substances are quite different, the measured spectra are almost identical. The angular dependence of the leakage spectra in H_2O was found to be very weak. The spectrum perpendicular to the surface has a mean energy about 17% higher than that of a Maxwellian. This difference decreases with increasing angle to the normal, being about 10% for grazing emission.

The measured perpendicular leakage spectra can satisfactorily be described by theoretical calculations made with ADALS. A modified version of the program for the Milne problem without absorption gives results which are in satisfactory agreement with the angular measurements. It is intended to improve this program by using still more terms in the variational trial function. We plan to make calculations for other scattering kernels, to see whether the angular dependence of the leakage is markedly influenced by variations in the scattering law.

III. Measurements of time dependent neutron spectra (W.R.)

1. Apparatus

The investigations of the time dependence of neutron spectra in zirconium hydride and ice were made with essentially the same equipment described above. For the zirconium hydride measurements we used a block of 20.0 x 26.0 x 28.0 cm of solid zirconium hydride ($ZrH_{1.65}$) which was in the form of small disc shaped pellets (15.1 mm diameter, 2.5 mm height) with a density of $\rho = 5.43$ g/cm^3. The average density of the block was 4.929 g/cm^3.

The ice was kept at low temperatures by a cryostat which is shown in fig.7. The ice block was of cylindrical form (14.2 cm diameter, 14.6 mm height) encased in a cadmium clad aluminium container, which was flanged to the storage vessel of the coolant. Through the small tube connecting the storage vessel with the moderator vessel, the coolant was in direct contact with the ice but had a negligible disturbing effect on the measurements. The temperature of the ice block was determined by two copper-constantan thermocouples. It was only slightly above the coolant temperature (77°K and 21°K respectively). The radiation shield, made of 1 mm copper, and the outer vessel had thin aluminium windows in order to minimize the absorption and scattering of neutrons in the beam.

2. Results

a) Zirconium hydride

Fig.8 shows the spectra in the middle of the zirconium hydride block at times after the fast neutron burst varying from 113 to 465 us. At the end of this time interval the spectra change only slightly with time, thus indicating that the asymptotic state has been approached. The spectra with the largest delay are cooler than a Maxwellian at moderator temperature (24°C) which can best be seen from fig. 9 , where the spectra divided by a Maxwellian of 20°C and corrected for the asymptotic decay $e^{-\lambda_0 t}$ are plotted versus flight time. This figure clearly shows the influence of the 0.13 ev resonance. Above 0.13 ev the neutrons are strongly scattered down to lower energies at early times which causes the sharp dip in the curves. At longer times, upscattering from lower

energies becomes important and, therefore, the spectra only change slowly in this region. The dip in the nearly asymptotic spectra is caused by diffusion cooling, since the moderator block was not infinitely large. The "constriction" of the curves occurs at 0.17 ev where the inelastic scattering cross section has a maximum.

From these spectra the time dependence of the mean energy has been determined. These data can be described well by an exponential approach of the mean energy to its equilibrium value with a relaxation time of τ_{rel} = 97 $\pm$ 7 μs (see fig.10). The asymptotic value of the mean energy is 0.0493 ev and thus 4% below 2 kT_M.

In addition, the spectra from the surface were measured from 50 μs to 370 μs after the pulse (fig.11). The almost asymptotic leakage spectrum is harder than the spectrum from the middle of the block by 18%. The mean energy of these spectra can again be fitted by an relation $\bar{E}(t) = \bar{E}_\infty (1 + a \, e^{\frac{-t}{\tau_{rel}}})$ with τ_{rel} = (74 $\pm$ 7) μs (fig.12).

b) Ice

The results of the spectra measurements in ice at 77°K are shown in fig.13. The spectrum measured with the longest delay after the neutron burst can be well described by a Maxwellian of 75°K, thus indicating a small decrease in mean energy by diffusion cooling. Close to the asymptotic state, the mean energy calculated from these data shows an exponential behaviour with a relaxation time τ_{rel} =(77 $\pm$ 7) μs (fig.14). Fig.15 shows the result of a transmission measurement through an indium foil, where Q(t) - Q(∞) is plotted versus time. (Q(t) is the ratio of the counting rate of the detector after transmission through the absorber to the counting rate of the bare detector.) The relaxation time obtained from these data is (67 $\pm$ 10) μs.

The results for ice at liquid hydrogen temperature given in figures 16 and 17 show that the moderation process at this low temperature is extremely slow. The dotted line is a Maxwellian of 21°K. From this, one can see that the neutrons are still far from their equilibrium state even at 744 μs after the pulse. At this time, the mean energy of the spectrum is about 30% higher than that of a Maxwellian of 21°K. In fig.18 the data of the spectra measurements are presented in the form $\emptyset(E,t)/M(E) \, e^{\lambda_0 t}$ (M(E) = Maxwellian of 21°K, λ_0 = asymptotic decay constant). During the moderation process a dip successively

appears in the curves in the vicinity of 0.01 ev. This phenomenon might be explained by a high density of states in the phonon energy distribution within this region. However, no confirmation for this could be found in the literature.

3. Discussion of the results and comparison with other measurements

a) The energy mode concept

For a long time it has been believed that the solution of the time dependent infinite medium diffusion equation without absorption can be decomposed into a set of discrete exponentially decaying energy modes

$$\emptyset(E,t) = \sum_{i=o}^{\infty} a_i \emptyset_i(E) e^{-\lambda_i t} . \tag{1}$$

Among others Corngold et al. [15] have shown that this concept is not true. Discrete eigenvalues exist only in the range $\lambda_i < (\Sigma_{inel} v)_{min}$ while above this limit the eigenvalues form a continuum. Therefore, the solution for the time dependent flux is written as

$$\emptyset(E,t) = \sum_{i=o}^{i_{max}} a_i \emptyset_i(E) e^{-\lambda_i t} + \int_{(\Sigma_{inel} v)_{min}}^{\infty} a(\lambda) \emptyset_\lambda(E) e^{-\lambda t} d\lambda \tag{2}$$

Under the condition that the two lowest eigenvalues are discrete, the approach to equilibrium is governed by the two first terms of the expansion

$$\emptyset(E,t) = \emptyset_o(E) e^{-\lambda_o t} + \emptyset_1(E) e^{-\lambda_1 t} \tag{3}$$

with
$$\lambda_o = 0$$

$$\lambda_1 = \frac{1}{\mathcal{T}_{th}} \qquad \mathcal{T}_{th} = \text{thermalization time} .$$

For a finite medium, where the spatial dependence can be taken into account by the geometric buckling B^2 , with $\frac{1}{v}$-absorption the same representation as eq.(2) holds with the single exception that the lower limit of the integral is given by $\lambda_{min} = (\Sigma_{inel} v)_{min} + \Sigma_a v.$

two cases eigenvalues are given by

$$\lambda_o = (\Sigma_a v)_o + DB^2 + O(B^4)$$

$$\lambda_1 = (\Sigma_a v)_o + \frac{1}{\widetilde{\ell}_{th}} + gDB^2 + O(B^4) \tag{4}$$

constant

factor.

the dependence of the mean energy is

$$\frac{\iint E e^{-\lambda_o t} + \int_o^\infty E\emptyset_1(E)dE \, e^{-\lambda_1 t}}{\iint E e^{-\lambda_o t} + \int_o^\infty \emptyset_1(E) \, dE \, e^{-\lambda_1 t}} = E_\infty \frac{1 + A e^{-(\lambda_1-\lambda_o)t}}{1 + B e^{-(\lambda_1-\lambda_o)t}}$$

$$= E_\infty \left((A-B) e^{-(\lambda_1-\lambda_o)t} + (B^2-AB) e^{-2(\lambda_1-\lambda_o)t} + \ldots \right) \tag{5}$$

Therefore, the determination of the relaxation constant of the mean

energy

$$\lambda_1 - \lambda_o = \frac{1}{\widetilde{\ell}_{th}} + (g-1) D B^2 + O(B^4) \ . \tag{6}$$

transmission measurements, where the counting rate of
the detector surrounded an absorber is divided by the counting rate of the
bare detector, leads a similar expression as the second line of eq.(5).

the truncation of the expansion after the second term
is jus should yield identical results.

b) De mination of v)_{min}

shap gold [16] have investigated the solution of the time
depend on for the two models of a heavy Debeye crystal and
a heav nstein crystal. They found that only for $\frac{\Theta_D}{T} \lesssim 0.28$ and $\frac{\Theta_E}{T} \lesssim 0.20$
discre values can obtained. This means that for practically no crys-
tallin um at perature a λ_1 value will exist.

Applying these results to ice of $T = 77^{\circ}K$ with an assumed Debeye temperature of $215^{\circ}K$ the value of $\frac{\Theta_D}{T} = 2.79$ is far beyond this limit. The same holds for zirconium hydride under the assumptions that it can be respresented by an Einstein crystal with an Einstein temperature which corresponds to 0.13 ev.

Unfortunately, little is known about the inelastic scattering cross sections for ice and zirconium hydride. Ehret [17] has done measurements on the inelastic scattering of zirconium hydride. The evaluation of the data has not yet been finished. Preliminary calculations of the total inelastic scatter-ing cross section with a width of 33 mev for the 0.13 ev resonance and a 2% contribution of the acoustical part of the frequency distribution yield a value of $(\Sigma_{inel}v)_{min} = 5.0 \cdot 10^4 \ sec^{-1}$ [18]. A calculation taking into account only the 0.13 ev resonance, assuming its width as 28 mev, and neglecting the acous-tical part gives $(\Sigma_{inel}v)_{min} = 2.5 \cdot 10^4 \ sec^{-1}$. These values correspond to re-laxation times of $20/us$ and $40/us$, respectively.

Therefore, the critical condition is well satisfied with the measured zirconium hydride values.

Recently, Whittemore [19] has published a new value of the width of 18 mev for the 0.13 ev resonance. This smaller value, however, will not appre-ciably change the inelastic scattering cross section at very low energies. Cal-culations, taking into account this new width, are being made.

For the determination of $(\Sigma_{inel}v)_{min}$ of ice at $77^{\circ}C$ the results of McReynolds and Whittemore [20] for the total cross section of ice have been used. The total cross sections of ice at $20^{\circ}C$ and $76^{\circ}K$ show a difference of about 10 barns at 0.0009 ev. Assuming that this difference is due to the inelastic scattering, which is essentially zero at $20^{\circ}K$, a value of $\sigma_{inel} = 10$ barns is obtained at 0.0009 ev. This energy is probably small enough that the $\frac{1}{v}$ - law for the inelastic scattering cross section is valid. From this value a limit-ing relaxation time of $40/us$ is obtained. The critical condition
$\lambda_1 - \lambda_o + DB^2 < (\Sigma_{inel}v)_{min}$ again is fulfilled.

c) Zirconium hydride

Under the assumptions that a discrete λ_1-value exists and that all higher energy modes had died out before the time at which the analysis of the data starts, the measured relaxation time constants can be identified with

$$\lambda_1 - \lambda_0 = \frac{1}{\tau_{th}} + (g-1) \, DB^2.$$

The agreement between the value of $74/\mu s$ obtained from the leakage spectra and that of $97/\mu s$ obtained from the spectra in the middle of the block is not very good. The difference may be due to the fact that higher energy modes are still present in the case of the leakage spectra. The time interval of the measurements begins at $50/\mu s$ after the pulse, whereas the analysis of the spectra from the middle starts at $113/\mu s$ after the pulse.

As the leakage from the block was small, these values will be close to the infinite medium values. Using the g-factor determined by Meadows and Whalen [21] we obtain

$$\tau_{th} = 92/\mu s \qquad \text{(middle of the block)}$$
$$\tau_{th} = 71/\mu s \qquad \text{(surface)} \ .$$

These data can be compared with the results of Meadows and Whalen [21]. Correcting our values for the difference in hydrogen density we get $126/\mu s$ and $98/\mu s$ for $ZrH_{1.7}$ with a density of $3.48 \ g/cm^3$, which are in reasonable agreement with values of Meadows and Whalen, obtained from the transmission measurement, of 139 and $124/\mu s$. The agreement with their thermalization time of $194/\mu s$ obtained from the lifetime measurements is not good.

d) Ice

For ice at liquid nitrogen temperature the relaxation times of $(67 \pm 10)/\mu s$ determined from the transmission measurement and $(77 \pm 7)/\mu s$ from the time behaviour of the mean energy agree well. As the leakage from the block was very small, these values are almost equal to the infinite medium values. Brown [22] has determined the thermalization time of ice at $76\,^{\circ}K$ by a transmission method to be $(20 \pm 2)/\mu s$ which differs very much from our data.

A rough estimate of the thermalization time can be made by means of the relation $\tilde{t}_{th} \approx \frac{6C}{D^2}$. With the diffusion parameters obtained by Antonov et al. [23], 136 μs are obtained. The above relation is only approximate and, therefore, the result gives only an idea of the order of magnitude for the thermalization time. It supports, however, our results which indicates that the energy exchange with the H_2O molecules is strongly hindered. In ice of liquid hydrogen temperature the mean energy does not show a purely exponential behaviour. This is to be expected as the neutrons are still far from equilibrium. If a first eigenvalue exists at all, a lower limit for the thermalization time can be obtained by the assumption that the region where the mean energy approaches its equilibrium value begins immediately after the end of the time interval of our measurements. This exponential approach is indicated by the dotted line in fig.18.from which a lower limit of about 800 μs is obtained.

4. Summary

The time dependence of neutron spectra in solid zirconium hydride of room temperature and ice at 77°K and 21°K have been investigated. From the mean energy of the spectra the values of $\frac{1}{\lambda_1 - \lambda_0}$ which for the infinite medium are identical with the thermalization time have been determined. The results are given in the following table:

	Buckling $B^2 \left[cm^{-2} \right]$	$\frac{1}{\lambda_1 - \lambda_0}$ $\left[\mu s \right]$	Remarks
$ZrH_{1.65}$ ($\bar{\rho}$ = 4.92 g/cm³)	0.0492	97 $\pm$ 7	
		74 $\pm$ 7	Leakage spectrum
Ice (77°K)	0.155	77 $\pm$ 7	
		67 $\pm$ 10	Transmission method
Ice (21°K)	0.155	> 800	

This analysis is based on the assumption that the concept of a thermalization time is reasonable and, if so, that it is possible to separate the two lowest energy modes from the contribution of higher modes. The results, however, are not accurate enough to state this with certainty. For zirconium hydride at 24°C and ice at 77°K a rough estimate indicates that the first condition might be fulfilled. The difference of the data for zirconium hydride show

that the contribution of higher modes might not be negligible. Because of these difficulties it seems more appropriate to compare the results of the spectra measurements directly with theory. Therefore, we intend to perform time--dependent spectra calculations for the investigated moderators.

Acknowledgements

References

[1]   BECKURTS, K.H., Z. Naturforsch. 16a (1961) 611.

[2]   KLADNIK, R. and KUŠČER, I., Nucl. Sci. and Eng. 11 (1961) 116.

[3]   WILLIAMS, M.M.R., Nucl. Sci. and Eng. 18 (1964) 260.

[4]   EISENHAUER, C., Nucl. Sci. and Eng. 19 (1964) 95.

[5]   KIEFHABER, E., Nucl. Sci. and Eng. 18 (1964) 404.

[6]   BARNARD, E. et al., BNL-719 (1962) 805.

[7]   BECKURTS, K.H. and REICHARDT, W., Proceedings of the EANDC Symposium
      on Neutron Time-of-Flight Methods, Saclay (1961) 239.

[8]   EYRICH, W., Nukleonik 4 (1962) 167.

[9]   GLÄSER, W. and BECKURTS, K.H., Nucl. Sci. and Eng. 20 (1964) 235.

[10] HAYWOOD, B.C. and THORSON, I.M., BNL 719, Vol.I (1962) 26.

[11] KLADNIK, R., NIJS Report-R 435, "The Asymptotic Angular Dependent
      Leakage Spectrum of Thermal Neutrons" (1964).

[12] KLADNIK, R., Nucl. Sci. and Eng 17 (1963) 185.

[13] FEDERIGHI, F.D. and GOLDMAN, D.T., KAPL-2225, "KERNEL and PAM-Programs
      for Use in the Calculation of the Thermal Scattering Matrix for Chemi-
      cally Bound Systems" (1962).

[14] MacDOUGALL, J.D., AEEW-M 318, "PIXSE", (1963).

[15] CORNGOLD, N., MICHAEL, P., and WOLLMANN, W., BNL 719 (C-32) Vol.IV
      (1962) 1103.

[16] SHAPIRO, C.S. and CORNGOLD, N., "The Approach to Equilibrium of a
      Neutron Gas", to be published in Phys. Rev.

[17] GLÄSER, W., EHRET, G., and MERKEL, A., Int. Symp. on the Inelastic
      Scattering of Neutrons, Bombay (1964), SM-58/15.

[18]   EHRET, G., private communication.

[19]   WHITTEMORE, W.L., GA-9744 (1964).

[20]   WHITTEMORE, W.L. and McREYNOLDS, A.W., GA-2503 (1960).

[21]   MEADOWS, J.W. and WHALEN, J.F., Nucl. Sci. and Eng. 13 (1962) 230.

[22]   BROWN, J.B., "Pulsed Neutron Source Measurements with Pure Moderators", Doctorate Thesis Birmingham (1963).

[23]   ANTONOV, A.V., et al., AEC-TR-5143.

Fig. 1

Set-up for measuring angular dependent leakage spectra
for θ values between 0° and 72°

Fig.2

Assembly for measuring the leakage spectra at the grazing angle

Fig.3

Perpendicular leakage spectra from a large sample at room temperature(24°C).
The solid curves are theoretical calculations.

I   : H_2O (•••); Maxwellian at moderator temperature (————)
II :   Dowtherm A
III: benzene
IV : H_2O (•••) compared to Dowtherm A (+++)
V : H_2O (•••) compared to benzene (+++)
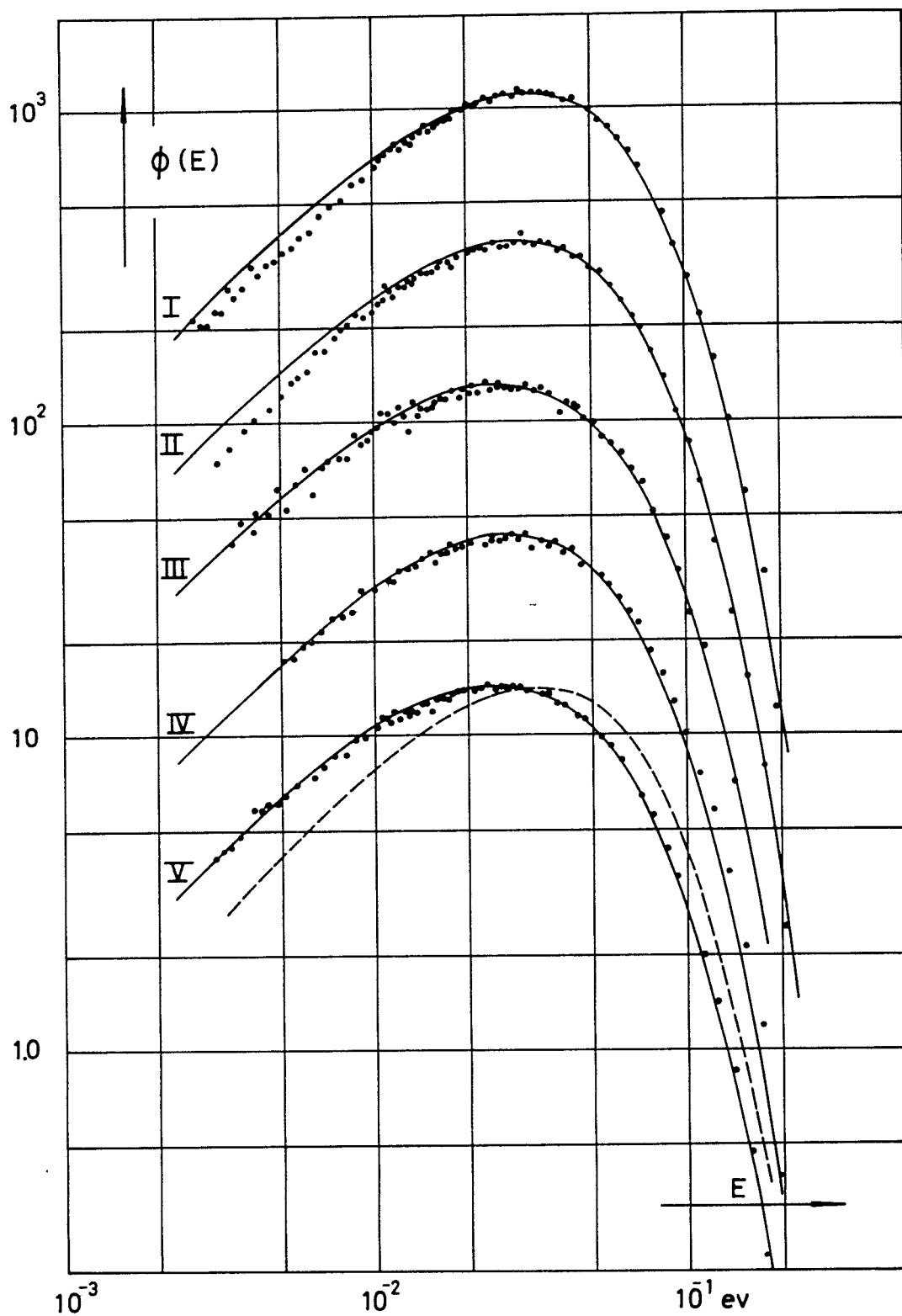
Fig.4

Perpendicular leakage spectra from thin slabs at 24°C.

The solid curves are theoretical calculations.

I   : H_2O, 4.2 x 30 x 30 cm slab
II : H_2O, 3 x 20 x 20 cm slab
III : H_2O, 2.1 x 20 x 20 cm slab
IV : benzene, 4.2 x 30 x 30 cm slab
V   : benzene (• • •), 3.1 x 30 x 30 slab;
      empirical curve fitted to points in fig.3.-IV (———)
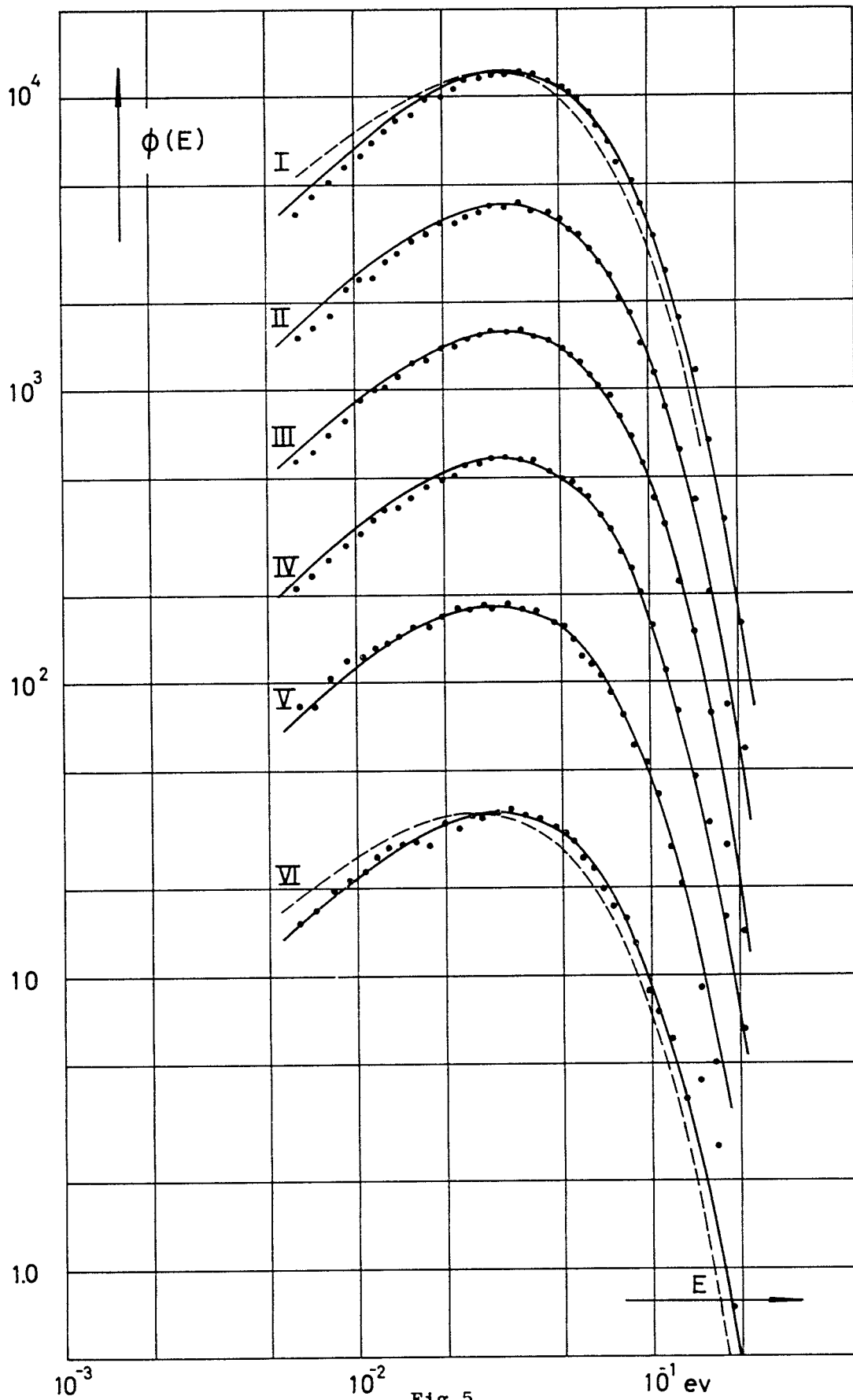
Fig.5

Angular dependent leakage spectra from H_2O at $24^{\circ}C$.
The solid curves are theoretical calculations.

I   : $\mu = 1.0$ ($\bullet\bullet\bullet$);
       empirical curve fitted to points in fig.5.-V (———)
II : $\mu = 0.7$ -   III: $\mu = 0.5$ -   IV : $\mu = 0.3$
V   : $\mu = 0.0$, extraction channel penetrating 4 mm below surface
VI : $\mu = 0.0$, extraction channel penetration 2 mm ($\bullet\bullet\bullet$)
                Maxwellian at moderator temperature (———)
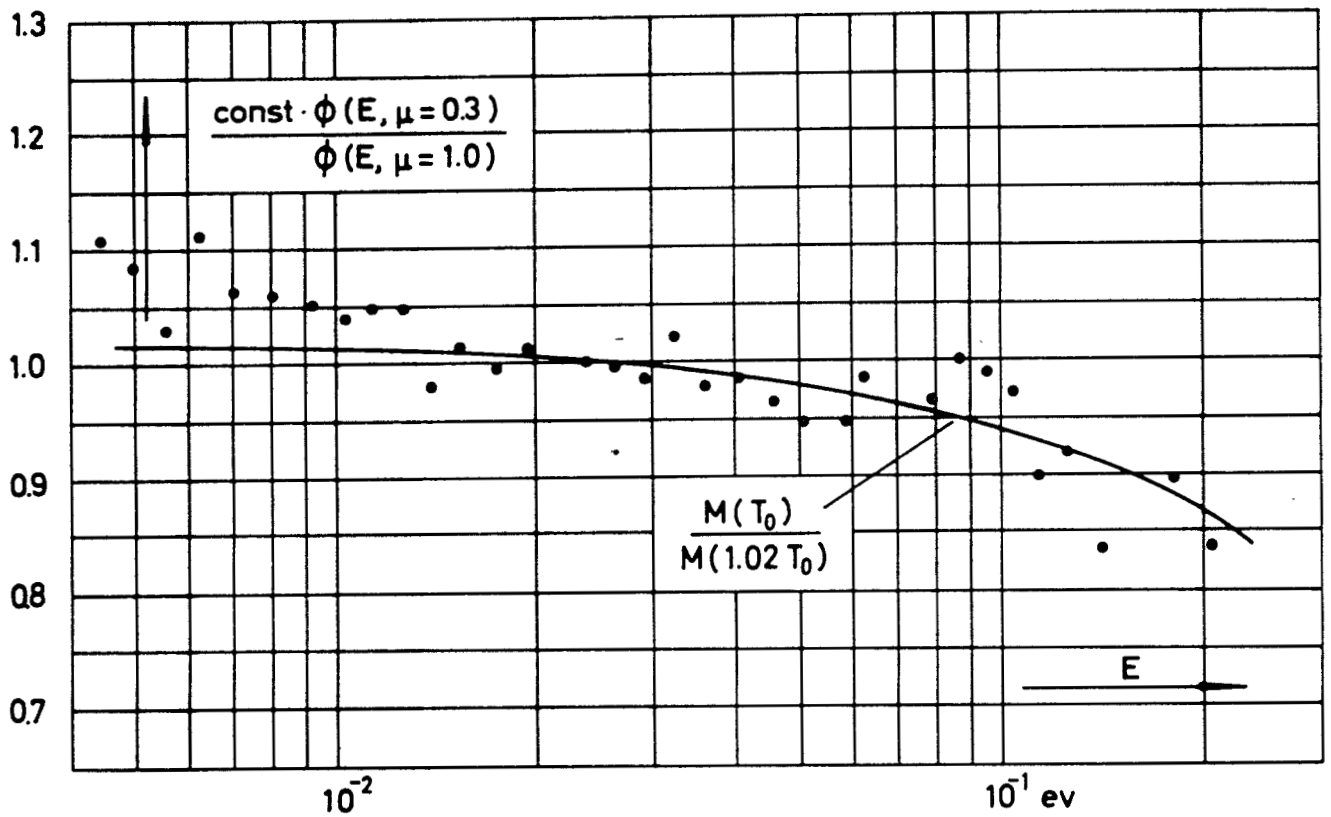
Fig.6

$\phi(E, u = 0.3)$ values divided by $\phi(E, u = 1.0)$ values.
The solid curve is the quotient of two Maxwellians with
T_o differing by 2 %.

liquid nitrogen
inlet

coolant
inlet

annular
liquid
nitrogen
tank

storage vessel
for the coolant

radiation
shield
(copper)

ice moderator

probe tube
for the BF$_3$-
counter for
life time
measurements

extraction channel
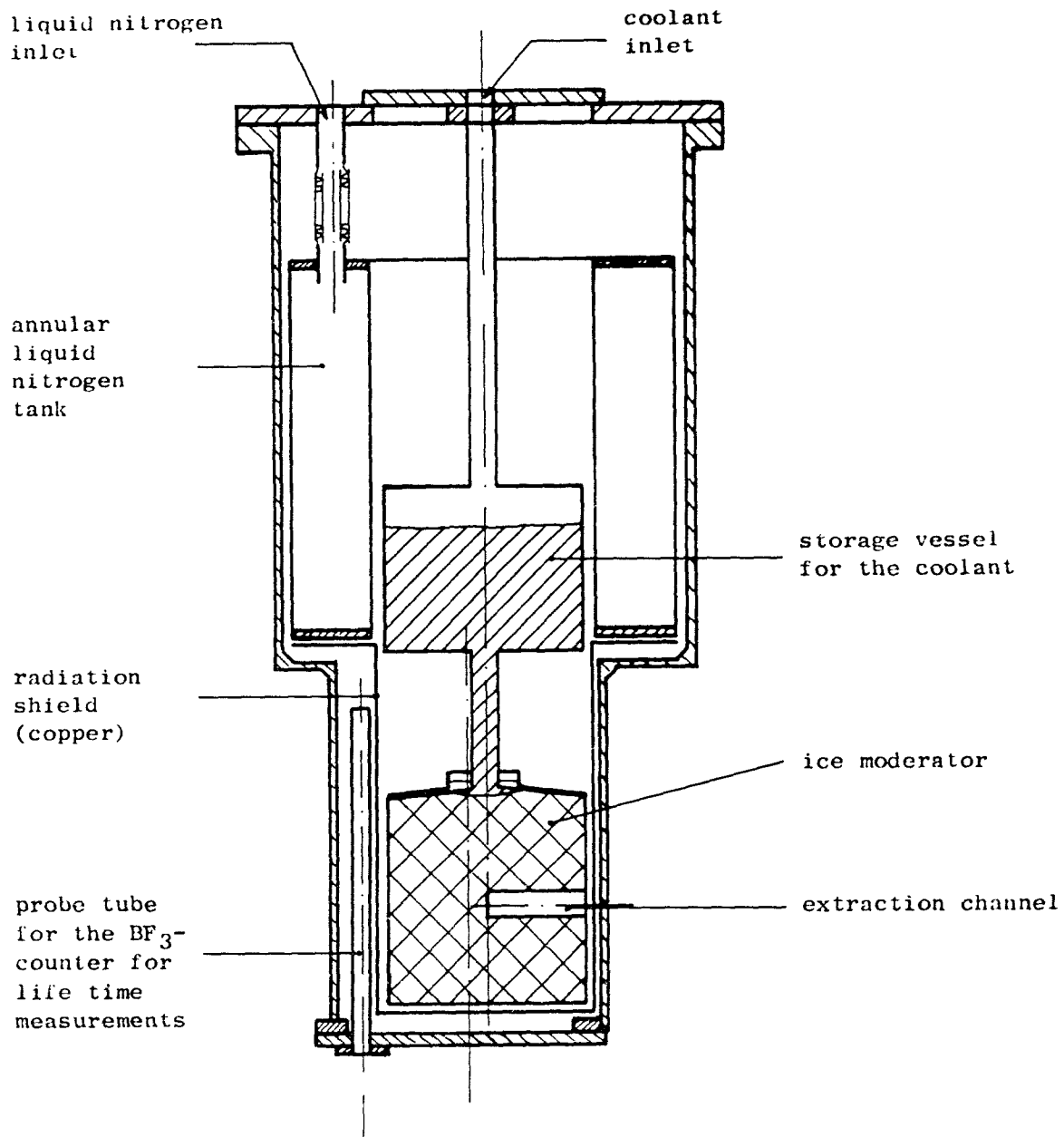
Fig.7

Schematic drawing of the cryostat
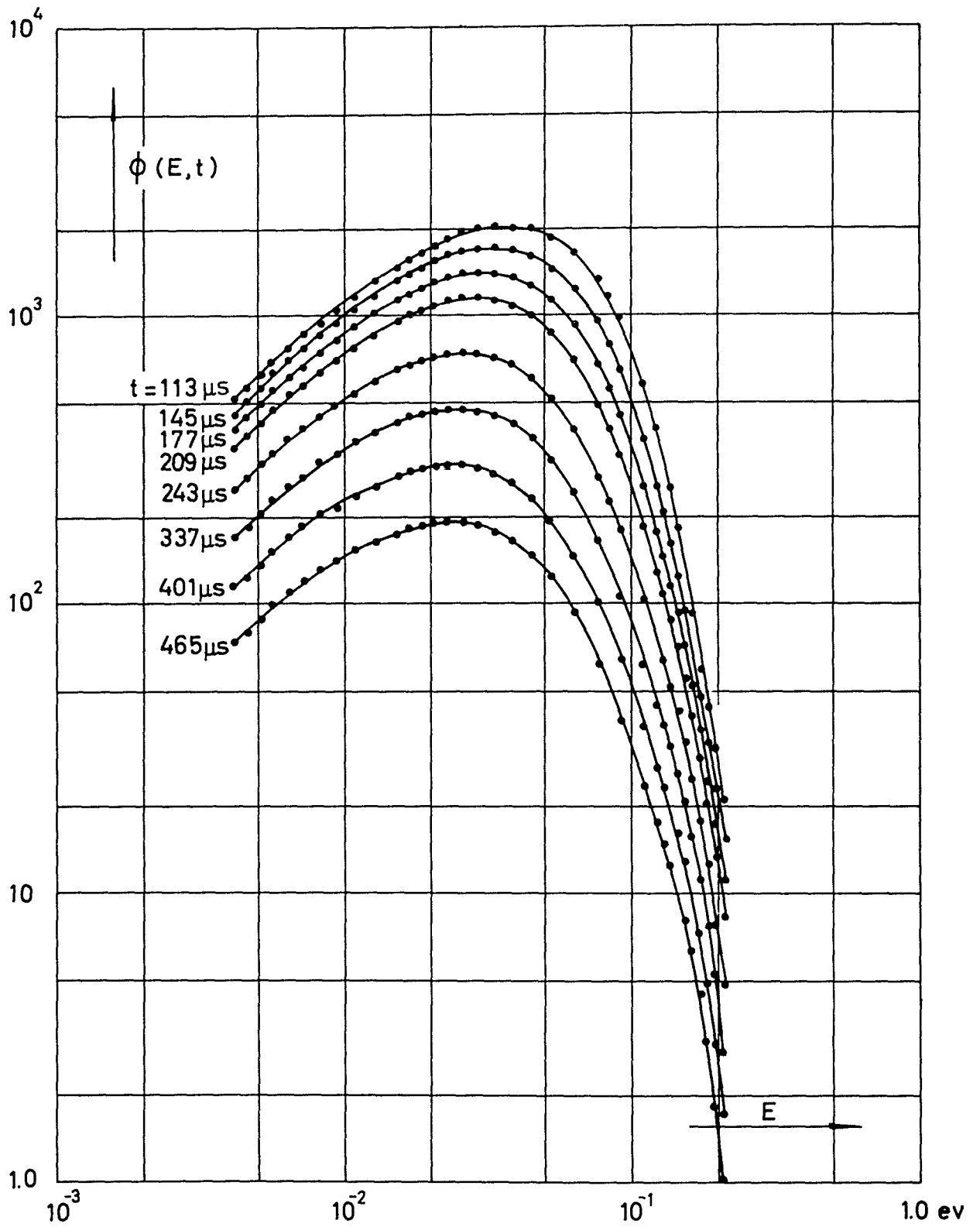
Fig.8

Spectra in zirconium hydride at different times after the neutron pulse
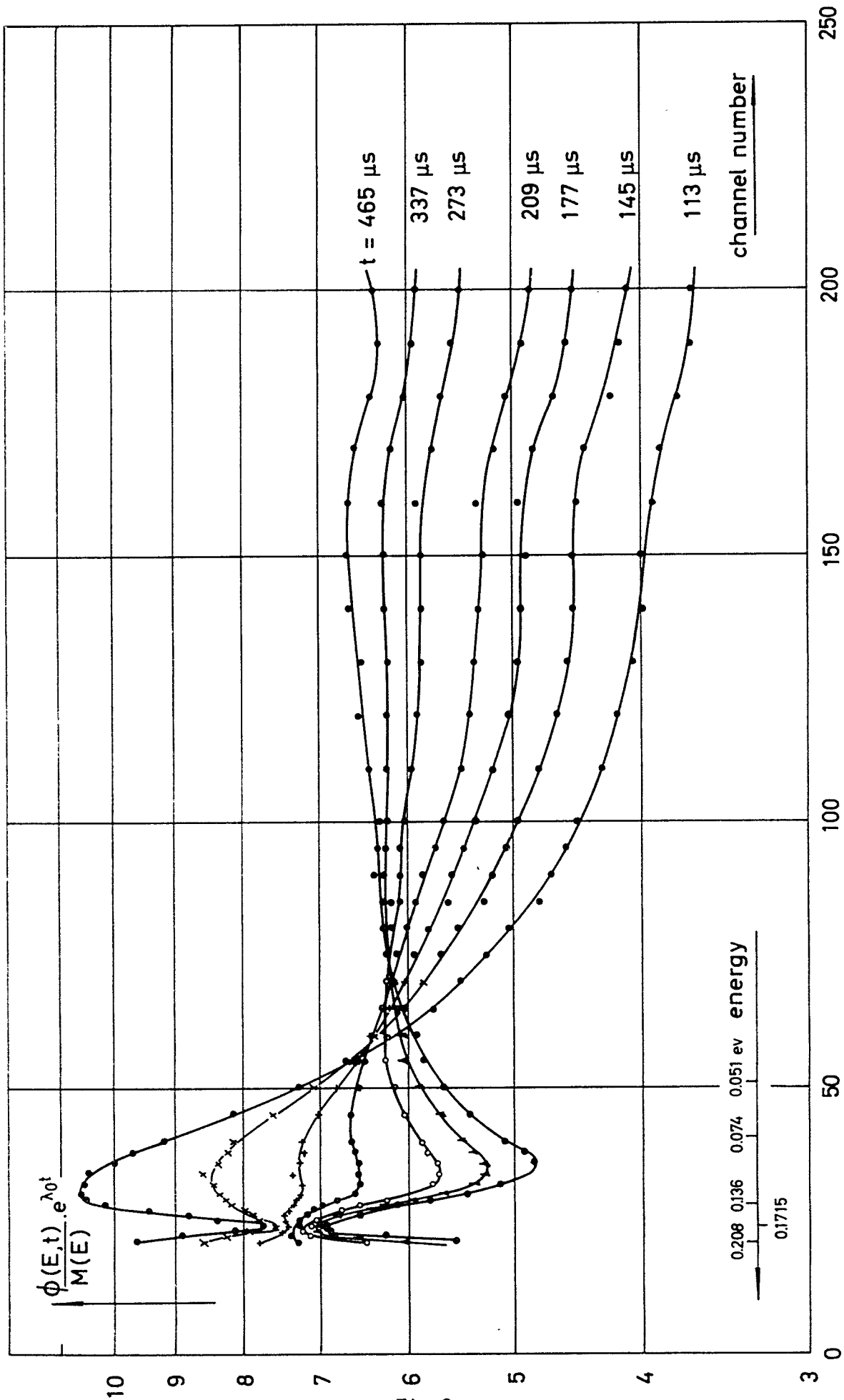
Fig.9

Spectra in zirconium hydride divided by a Maxwellian of 20°C
and corrected for the asymptotic decay versus flight time

- 25 -
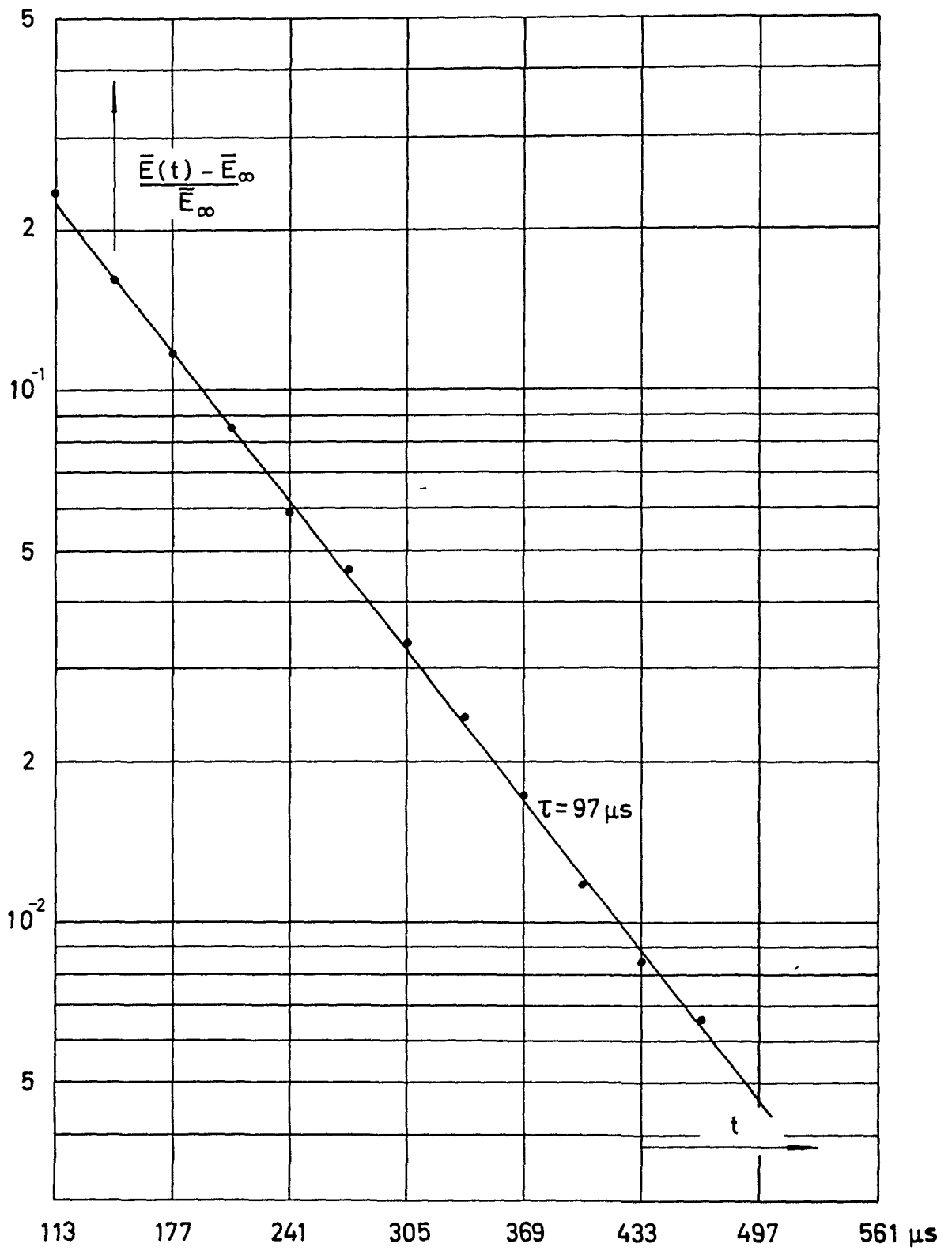
Fig.10

Determination of the relaxation time of the mean energy in zirconium hydride
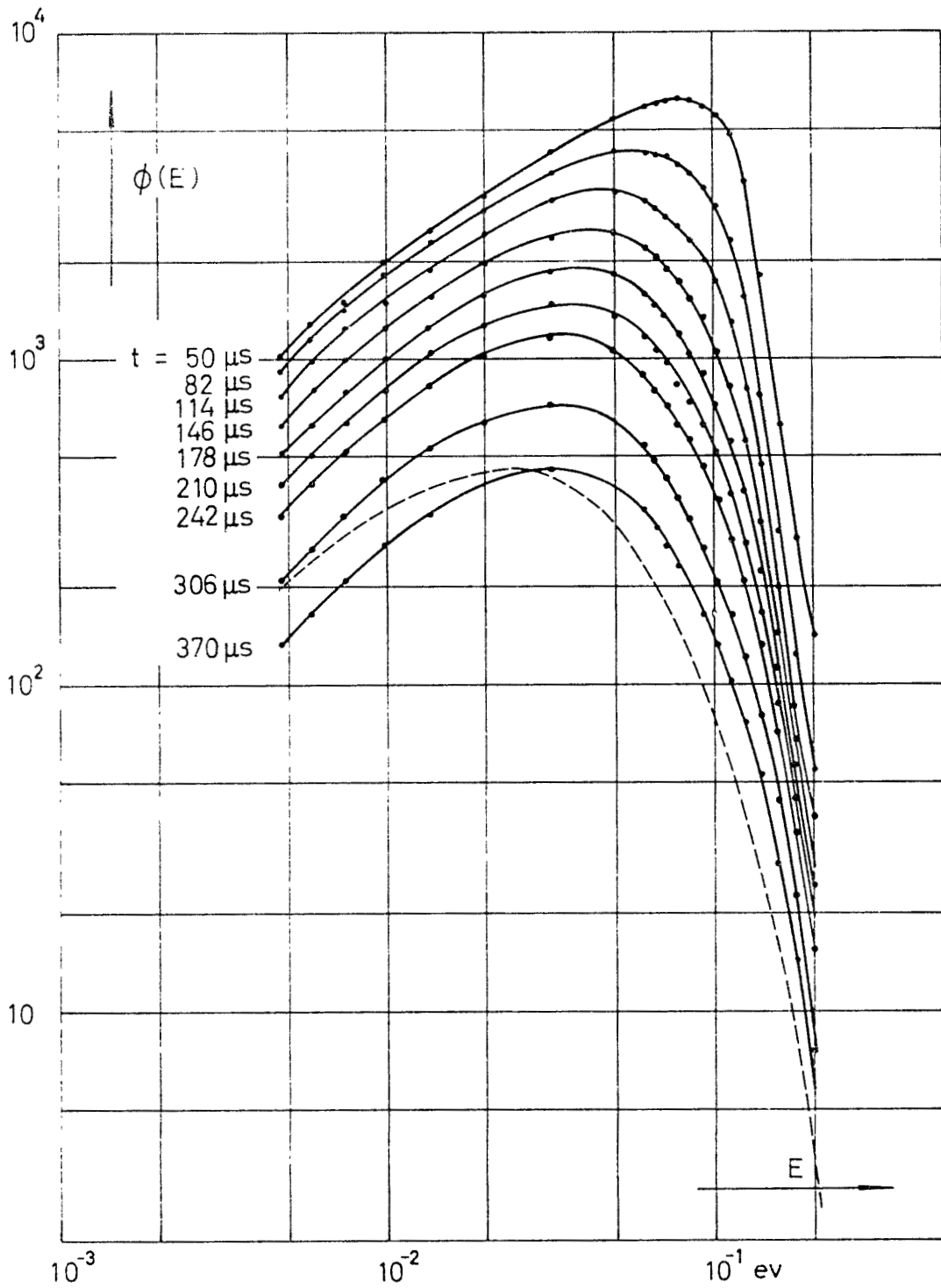
Fig.11

Leakage spectra from zirconium hydride
at different times after the neutron pulse

$$\bar{E}(t) - \bar{E}_\infty$$

10

1.0

$\tau = 74\,\mu s$

t

50   114   178   242   306   370   434 μs
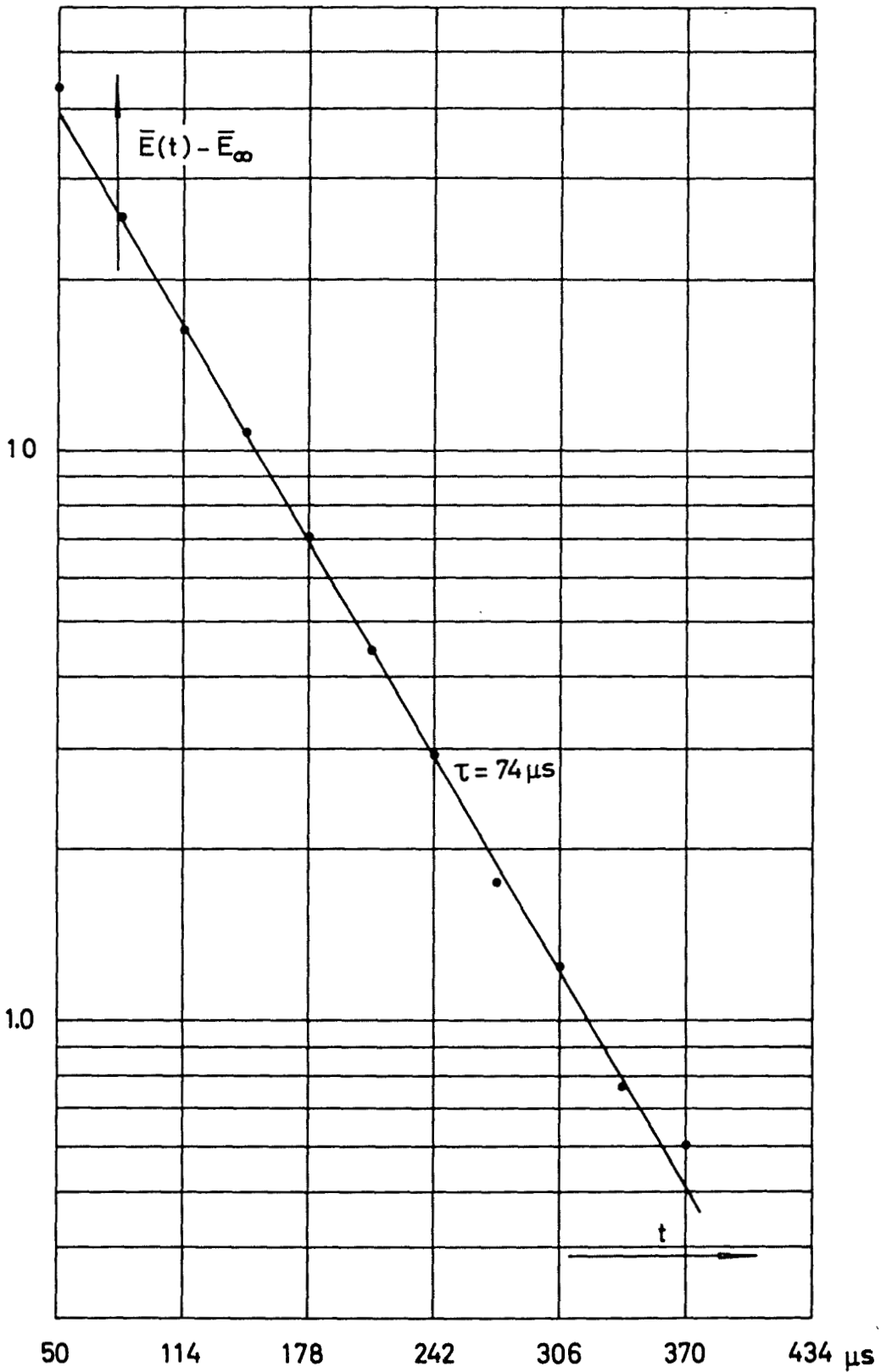
Fig.12

Determination of the relaxation time of neutron energy
of the leakage spectra from zirconium hydride
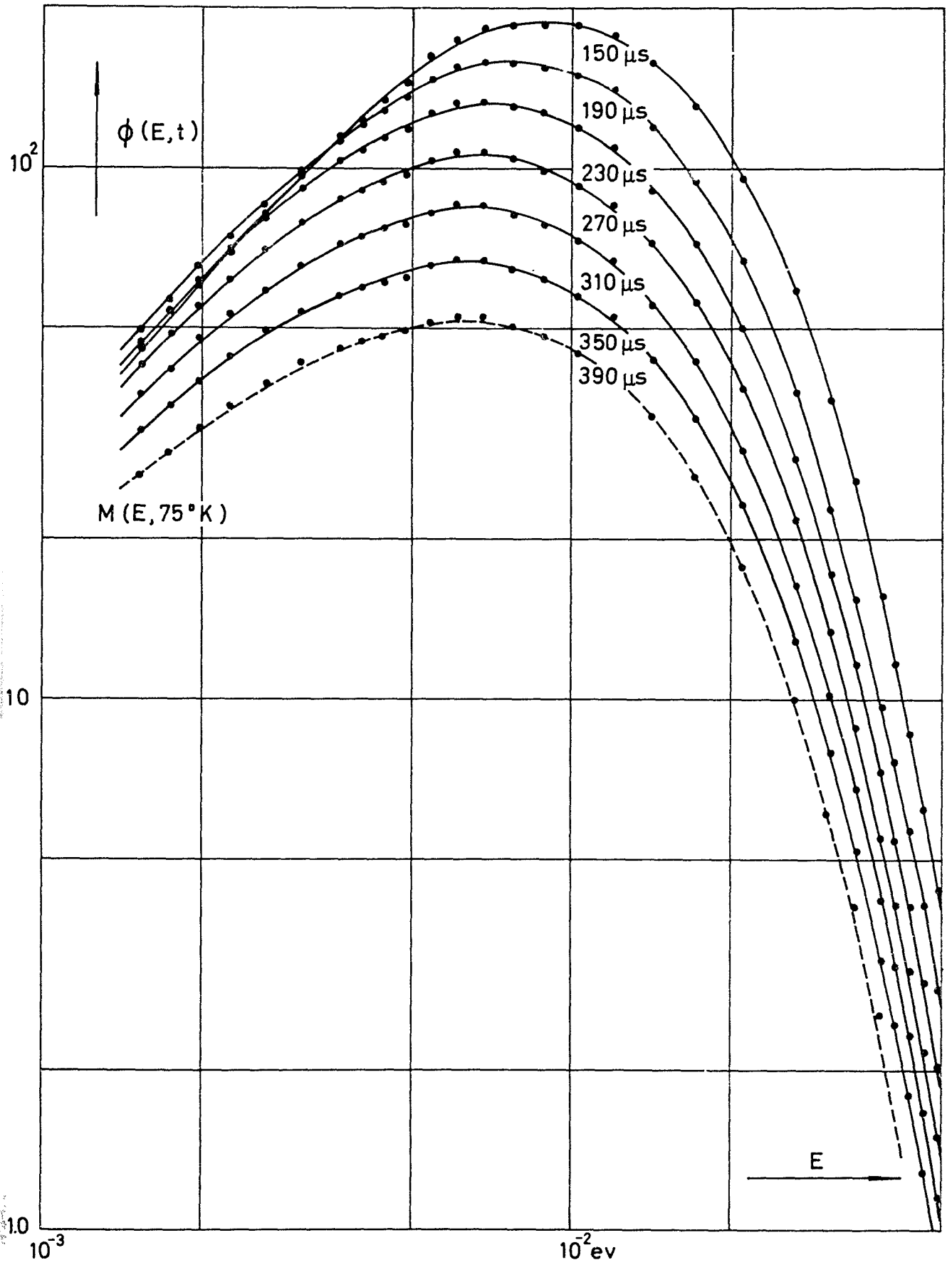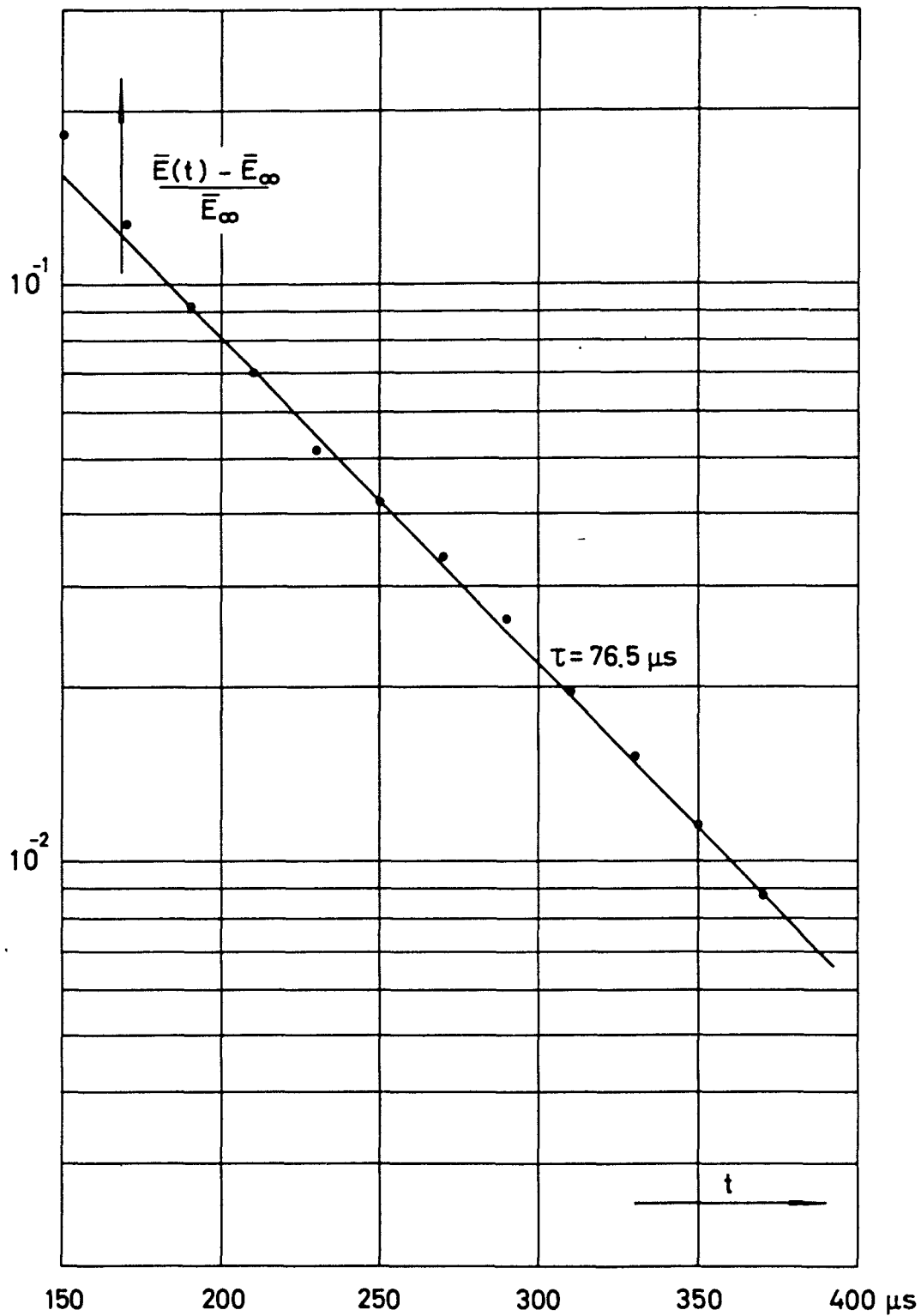
Fig.13

Time-dependent neutron spectra in ice at 77°K

$$\frac{\bar{E}(t) - \bar{E}_\infty}{\bar{E}_\infty}$$

$\tau = 76.5\ \mu s$

t

150   200   250   300   350   400 μs

Fig.14

Time dependence of the mean energy in ice at $77^O K$
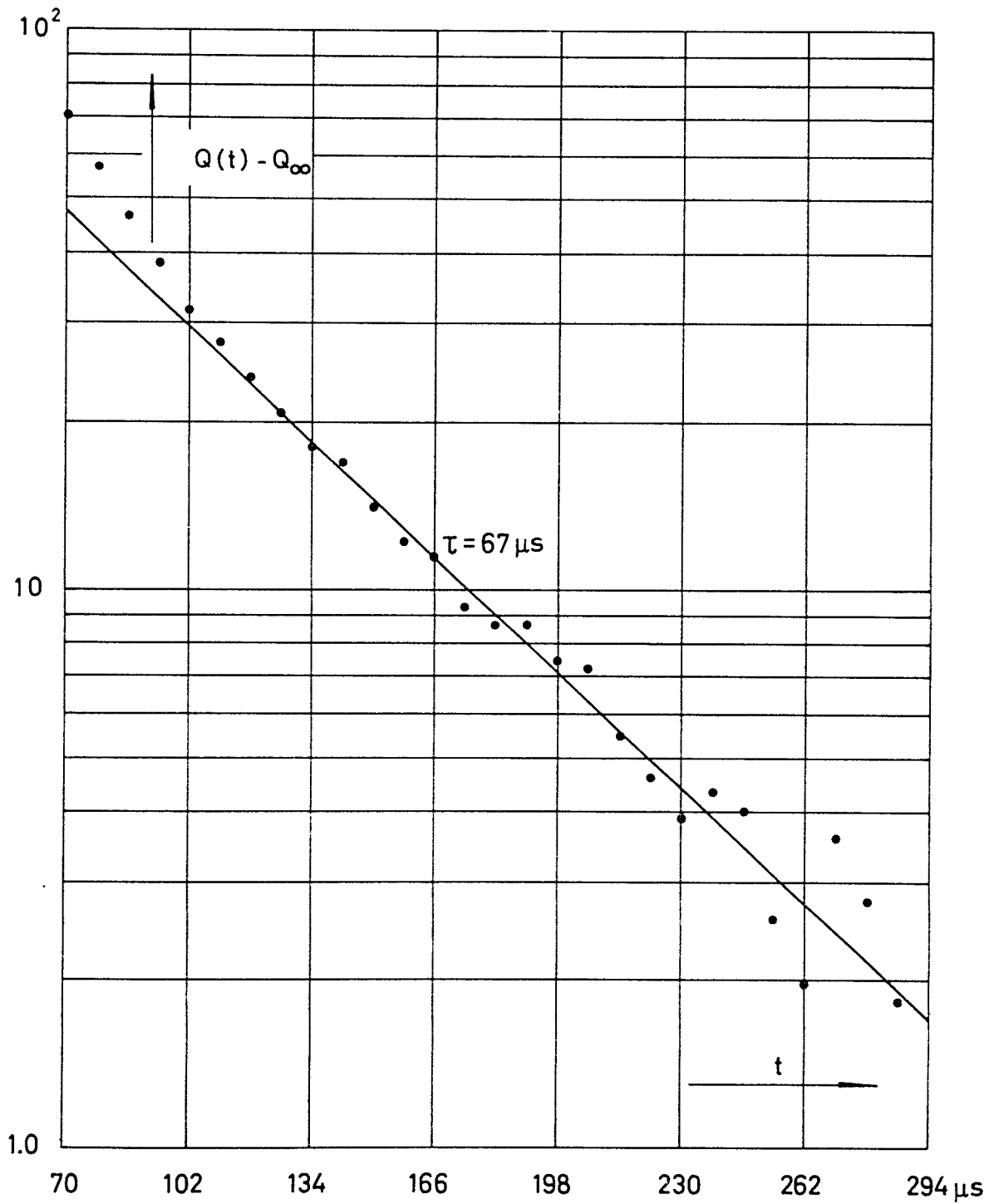
Fig.15

Ice at 77°K: Determination of the relaxation time from a transmission measurement
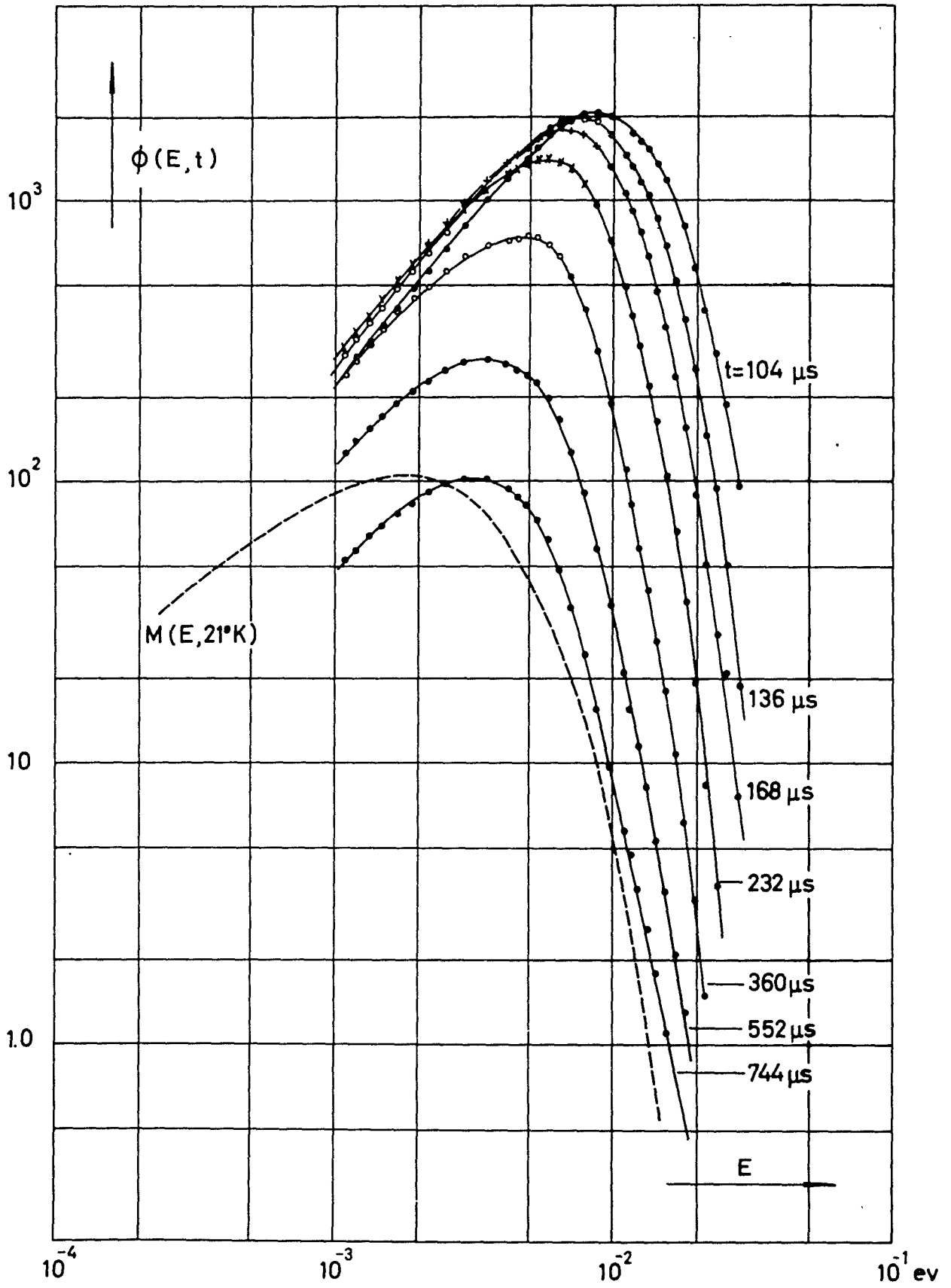
Fig.16

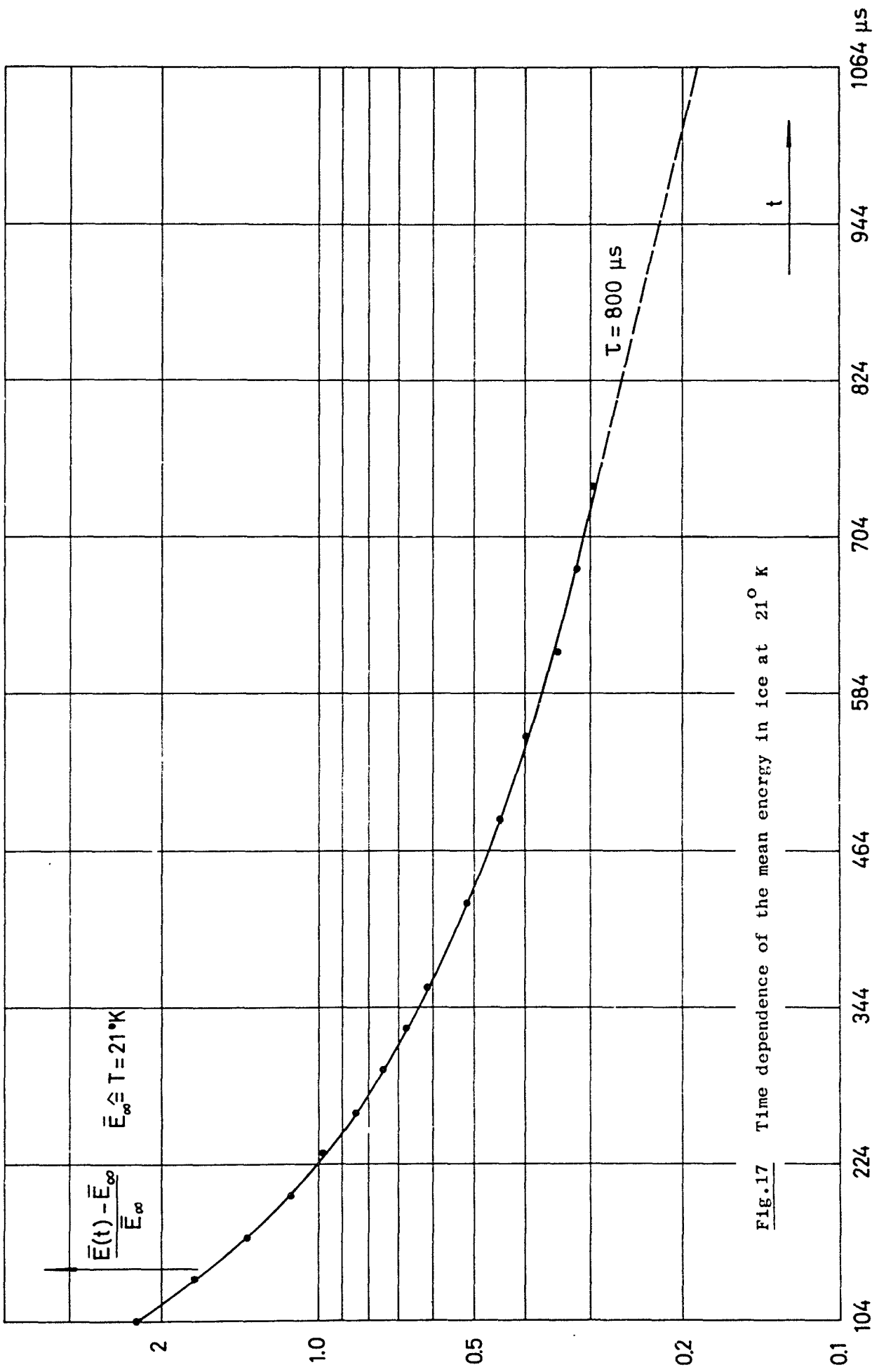Neutron spectra in the ice at 21°K at different times
after the neutron pulse
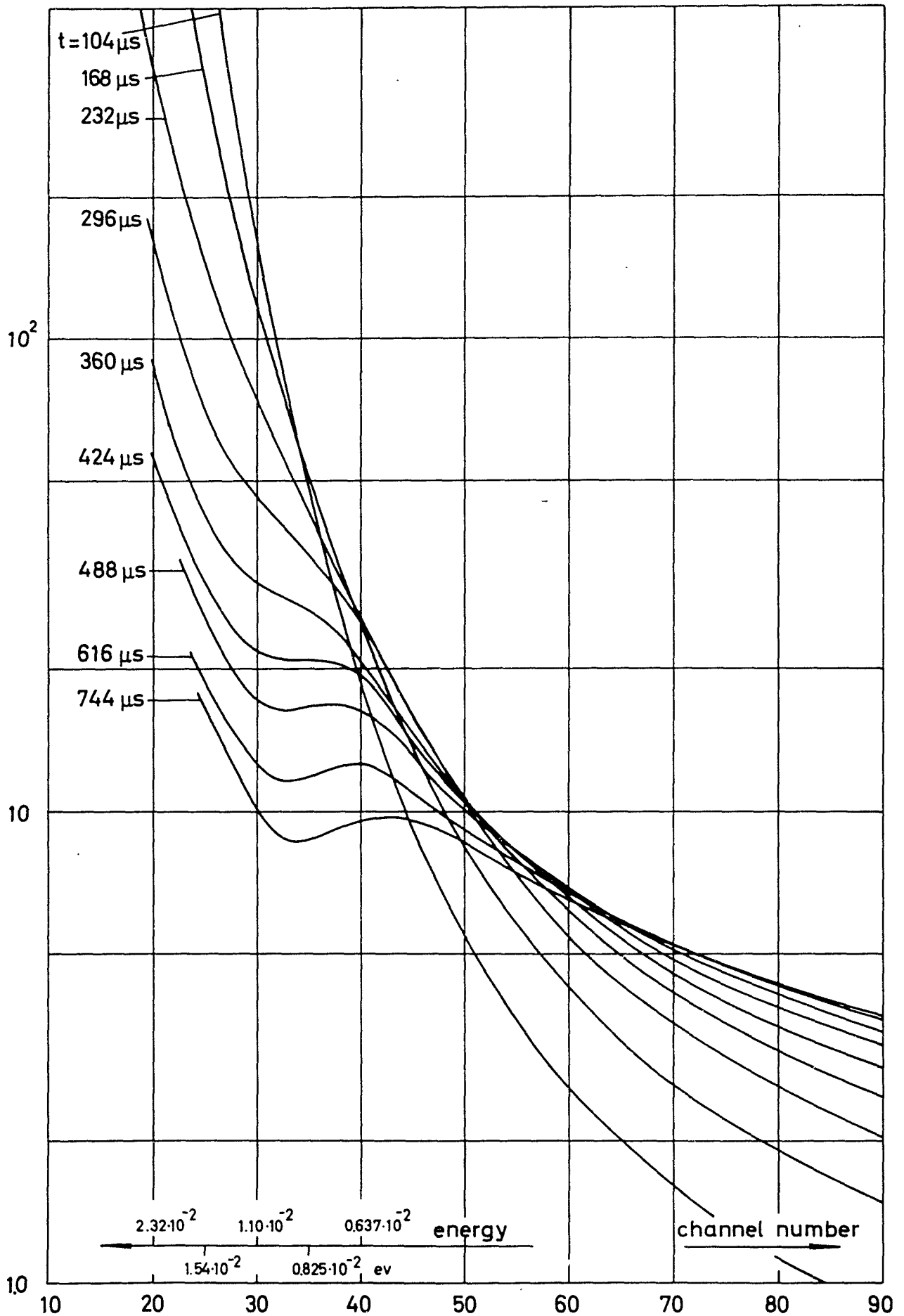
Fig.17 Time dependence of the mean energy in ice at 21° K

Fig.18

Ice at $21^{\circ}K$: $\dfrac{\phi(E,t)\ e^{\lambda_0 t}}{M(E,T=21^{\circ}K)}$ versus flight time