A. Ritchie

APPROVED FOR PUBLICATION (22) AAEC/TM456

AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT
LUCAS HEIGHTS

MEASUREMENTS IN PULSED BeO ASSEMBLIES WITH DECAY CONSTANTS IN THE REGION OF CORNGOLD'S LIMIT

by

M.T. RAINBOW

A.I.M. RITCHIE

May 1968

APPROVED FOR PUBLICATION

AUSTRALIAN ATOMIC ENERGY COMMISSION

RESEARCH ESTABLISHMENT

LUCAS HEIGHTS

MEASUREMENTS IN PULSED BeO ASSEMBLIES WITH DECAY CONSTANTS IN THE REGION OF CORNGOLD'S LIMIT

by

M. T. RAINBOW*

A. I. M. RITCHIE

ABSTRACT

Decay curves have been measured in BeO assemblies with bucklings in the range 1.93×10^{-2} cm^{-2} to 5.34×10^{-2} cm^{-2} at times from immediately after the initial pulse to at least 4.5 ms after the pulse. The decay curves for assemblies with $B^2 < 2.91 \times 10^{-2}$ cm^{-2} can be described by an exponential, from a minimum of 1.5 ms after the pulse to greater than 3.0 ms after the pulse. For bucklings greater than $B^2 = 3.3 \times 10^{-2}$ cm^{-2} the decay curves cannot be described by an exponential in this time region, and the departure from exponential increases with increasing buckling.

The results indicate a transition region at $B^2 \sim 3.0 \times 10^{-2}$ cm^{-2}, ($\lambda \sim 3.65 \times 10^{3}$ sec^{-1}) which is consistent with the expression proposed by Kothari for the upper limit on discrete decay constants and well above Corngold's limit of

Continued...

* University of Tasmania, Hobart.

ABSTRACT (Continued)

$\lambda^* = (v\,\Sigma_{inel})_{min} \sim 2.5 \times 10^3\ sec^{-1}$. The transition point does not have any marked shape dependence. There is some shape dependence in the behaviour of decay curves for assemblies with $B^2 \geq 3.3 \times 10^{-2} cm^{-2}$ but little dependence on pulse length, or counter shape and position.

NOTE:

This work has been submitted to a journal. Further details can be obtained from the authors or from the Director of the Research Establishment.

CONTENTS

1. INTRODUCTION

Since the 1962 BNL Conference on Neutron Thermalisation a number of theoretical papers have appeared (Corngold 1964, Albertoni and Montagnini 1965, Bednarz 1965, Kuscer and Corngold 1965) on the spectrum of the operator appearing in the Boltzmann equation. Albertoni and Montagnini have shown that for a gas kernel the spectrum consists of two parts: a set of points on the real axis, bounded on the left by the line Real $(\lambda) = -(v\Sigma)_{min}$ and belonging to the point spectrum, and the half-plane to the left of the line Real $(\lambda) = -(v\Sigma)_{min}$ which belongs either to the continuous or residual spectrum. As the size of the system decreases the point spectrum moves to the left until, for a sufficiently small system (mean chord ~ mean free path), the point spectrum vanishes. The theorems of Functional Analysis cannot easily be applied to the kernels for liquids and polycrystalline materials because of the elastic components in the scattering cross section of these moderators. Hence rigorous statements cannot be made about the spectrum of the operator appearing in the Boltzmann equation when these kernels are used. Moreover, the large discontinuity at the Bragg cut-off in the transport cross section of polycrystalline materials further complicates the problem.

Corngold has indicated that in an infinite liquid or polycrystalline assembly the point spectrum is bounded by $(v\Sigma_{inel})_{min}$. Since as $v \to 0$ all inelastic scattering cross sections vary as $1/v$ and swamp the elastic cross section, this bound is effectively the same as $(v\Sigma)_{min}$. The same author has shown that if diffusion theory is applied to a finite medium the bound is replaced by

$$(v\Sigma_{inel} + \tfrac{1}{3} v \lambda_{tr} B^2)_{min} , \qquad .....1$$

which exhibits a number of maxima and minima in a polycrystalline material due to the effects of the various Bragg peaks, but is monotonically decreasing in a liquid. However, in the common polycrystalline moderators the minimum still occurs at $v = 0$.

Table 1 shows some values for $(v\Sigma)_{min}$ in gases and in common liquid and polycrystalline moderators. It can be seen that the limiting decay constant in water or heavy water systems is well beyond the normal experimental values encountered but that the limiting values for polycrystalline materials are within the region normally used for the pulsed type of experiment. In fact some workers (Iyengar 1957, Andrews 1960, Zhezherun 1964, Joshi et al. 1967) have reported $\lambda(B^2)$ curves for polycrystalline moderators containing points with decay constants

significantly greater than $(v\Sigma_{inel})_{min}$. In an effort to explain this discrepancy Kothari (1965) points out that there will be very few neutrons of low energy ($\leq 10^{-4}$ eV) in any assembly so that it is physically unrealistic to consider the absolute minimum of the expression 1. He claims it is more physically realistic to find the minimum of this function in the energy region of maximum neutron density. This minimum will occur at the maximum of the transport cross section and the value of the decay constant will then be limited by

$$(v\Sigma_{inel} + \tfrac{1}{3} v\lambda_{tr} B^2)_{v = v_o} \qquad \ldots\ldots 2$$

where v_o is the velocity corresponding to the maximum transport cross section.

However, since $(v\Sigma_{inel})_{min}$ does represent the minimum neutron reaction rate and since inelastic scatter in the region of the Bragg cut-off is non-zero, neutrons described by the continuous mode will eventually decay with the decay constant $(v\Sigma_{inel})_{min}$. Kothari claims that this will take a long time to happen, (~ 5 to 10 ms) an argument similar to that used by de Saussure (1962) and Jha (1960), in describing the 'trapping effect'.

The absence of an asymptotic decay mode has been demonstrated experimentally by Gaerttner et al. (1965) and Silver (1962) in beryllium. However, the buckling of 7×10^{-2} cm^{-2} used by Gaerttner et al. and the temperature of ~ -96°C used by Silver both lead to decay constants so far from the continuum limit that the theories of both Corngold and Kothari predict a non-asymptotic mode of decay. More recently Ritchie and Rainbow (1967) have shown that in small beryllium oxide assemblies decay curves exist which can be described by an exponential over a period of about 2.0 to 3.5 ms after the primary neutron pulse, even though the decay constants are greater than $\lambda^* = (v\Sigma_{inel})_{min} \simeq (2.5 \times 10^3\ sec^{-1})$. However, for bucklings greater than 2.92×10^{-3} cm^{-2} ($\rho = 2.96$ g cm^{-3}), which corresponds to a decay constant of 3.7×10^3 sec^{-1}, the decay cannot be described by an exponential. These results do not agree with Corngold's prediction but are consistent with Kothari's. Zhezherun (1967) has carried out measurements in beryllium and beryllium oxide assemblies. He also finds decay constants greater than λ^* (~ 3.8×10^3 sec^{-1} for Be) and consistency with Kothari's limit. However, in the case of beryllium oxide he evaluates Kothari's expression at the second major peak in the transport cross section (0.016 eV) since he claims that because of extinction effects this will give, in practice, a higher transport cross section than the peak at 0.005 eV.

Zhezherun finds that for beryllium oxide assemblies with bucklings as high as 5.6×10^{-2} cm^{-2} ($\rho = 2.96$ g cm^{-3}) the decay curves are exponential. This disagrees with the work of Ritchie and Rainbow (1967) but it should be noted that he measures the decay curves only over the time range 0 to 2.0 ms with waiting times of no longer than 1.12 ms. More recently Joshi et al. (1967) has measured decay curves which are exponential at buckling values of 5.97×10^{-2} cm^{-2} but their waiting times are even shorter ($\sim$ 500 μs) than those of Zhezherun.

In an attempt to clear up this disagreement and to investigate the apparent consistency of Kothari's limit with experimental observations, further experiments have been done with small beryllium oxide assemblies. The present measurements cover the buckling range 1.93×10^{-2} cm^{-2} to 5.34×10^{-2} cm^{-2} which corresponds to an absorption-corrected decay constant range from 2.43×10^{3} sec^{-1} (well below $(v\Sigma_{inel})_{min}$) to well above Kothari's limit. In each case the decay curve has been measured from immediately after the pulse till at least 4.5 ms after the pulse and in most cases covers a total of 8 decades of count rate. Various shapes of assemblies were used to investigate the usefulness of the buckling concept in small assemblies and the effect of shape on the limiting values of the decay constant. The effect of source conditions, counter position and counter size were also investigated since for an assembly with non-exponential behaviour the decay described by a continuous mode of decay should be much more source-dependent than one which can eventually be described by an asymptotic mode. The results of these measurements are given in Section 4 and discussed in Section 5.

For decay curves not described by an exponential an attempt has been made to ascribe 'effective' decay constants at 1.0, 1.5, 2.0 and 2.5 ms after the pulse, and plot these as a function of buckling. The results of this approach are given in Section 4 and their relationship to the measurements of Zhezherun and other workers is discussed in Section 5.

2. APPARATUS

2.1 Counting Equipment

The time analyser used was the modified Laben 512-channel analyser described previously (Ritchie 1967) and operated throughout the present measurements with 64 channels. This gave a fair compromise between good time resolution and good counting statistics. In order to improve the count rate at long times after the pulse, two 12EB40 BF_3 detectors were used in parallel. The outputs of the

detectors passed through parallel amplifiers (AERE Type 1430A) whose outputs were fed into the time analyser through a common discriminator. A block diagram of the electronics is shown in Figure 1.

2.2 Beryllium Oxide Assembly

The assembly and shielding are essentially the same as that used previously in the $\lambda(B^2)$ measurements (Ritchie 1967) and are shown schematically in Figure 2. The beryllium oxide (the same sample as used in the $\lambda(B^2)$ measurement) was in the form of sintered tiles 15.24 x 15.24 x 2.54 cm^3 of average density 2.867 g cm^{-3}. The pulsed neutron source was a modified (Fraser et al. 1968) 3 MeV Van de Graaff accelerator which produced pulses at intervals of between 5 and 10 ms, depending on the assembly under study, and was operated throughout most of the experiment with a pulse length of 500 μs. The $^9Be(d,n)^{10}B$ reaction was used and, with up to 700 μA of peak current, provided a copious source of neutrons. It was found that the background level varied slowly from run to run. This was attributed to the fact that the d.c. level in the pulse varied between 0.01 per cent and 0.2 per cent depending on the age and condition of the ion source, and the quantity of beam being extracted. Although this d.c. current was deflected from the target in the off-pulse period by the post-acceleration deflector system, neutrons produced by the $D(d,n)^3He$ reaction at the point of contact of the deflected beam with the flight tube wall, passed down the flight tube and into the beryllium oxide assembly, so causing a background.

Since the object of these measurements is to note a departure from the exponential mode of decay it is important that the decay of the assembly is divorced from the decay of any materials around the assembly. The borated paraffin and boral shielding arrangement around the assembly proved sufficient in the $\lambda(B^2)$ experiment (Ritchie 1967) to make the decay constant of any assembly independent of changes in massive concrete shielding in the vicinity of the stack. As a further check a measurement was done with all the beryllium oxide removed from the assembly and the Van de Graaff accelerator producing pulses 500 μs long every 5 ms with 600 μA peak current. The BF_3 counters were placed at the centre of the assembly and in a position similar to that used when measuring the decay of a given beryllium oxide assembly. The result of such a measurement in which the data collection time was some 70 min is shown in Figure 3 where it can be seen that there is no time dependence in the background ~ 800 μs after the pulse.

3. EXPERIMENTAL TECHNIQUE

3.1 Measurement of Decay Curves

For each stack size the table on which the assembly rested was adjusted to bring the source into line with the centre of the front face of the assembly. This eliminated asymmetry except in the beam direction. The two detectors were held in a cadmium holder fitted with a slot 5 x 10 cm^2 designed to define the spatial region exposed to the counters. In all cases where an attempt was made to measure a decay constant the detectors were placed on the top surface of the beryllium oxide with their length parallel to the beam direction and in all but two cases with the centre of the active volume at the centre of the stack. The two exceptions were the 60.96 x 45.72 x 15.24 cm^3 and the 30.48 x 30.48 x 15.24 cm^3 stacks where the boral shielding around these assemblies made it necessary to shift the centre of the sensitive volume of the counter about 1.0 cm from the centre of the stack in a direction away from the source.

It was intended, in this investigation, to measure for each assembly the whole of the decay curve from immediately after the initial neutron pulse until at least 4.5 ms after the pulse. However, because of dead time considerations and the limited storage (65,536) of the time analyser used it was impossible to do this in one measurement. Consequently, the time range was divided arbitrarily into three overlapping regions which covered the following approximate periods:

I - 0 to 2 ms after the pulse

II - 1 to 3 ms after the pulse

III - 2 to $\geq$ 4.5 ms after the pulse

The decay curve was measured for each assembly in Region III first of all. The maximum source intensity was used and this run continued until the maximum storage had been reached in the channel corresponding to about 2 ms after the pulse. These runs were repeated until a total count of about 10^6 had been recorded at 2 ms. The source strength was reduced by about a factor of ten to give ~ 1 per cent dead-time correction at 1 ms and several runs done to obtain a total of ~ 10^6 counts at 1 ms. A further reduction by a factor of ten in source strength allowed measurements to be done in Region I where again about 10^6 counts were obtained immediately after the pulse. If the decay were exponential over the whole time region this would allow it to build up over the full 5 ms. In general it is not so, but each region can be analysed to note any systematic changes in the decay constants within each region and from region to region.

3.2 Data Analysis

The data from each time region was analysed by the code EXPFIT, which used the weighted least sum of squares criterion to determine the parameters giving the best fit to the data. The Normal Equations derived from the least squares criterion are non-linear and were solved by an iteration method in which the equations were linearised using a Taylor expansion about estimates for the parameters. Two forms of EXPFIT were used:

(i) EXPFIT (1), which fitted data to the form $A_1 + A_2 e^{-\lambda t}$ where the background A_1, amplitude A_2 and decay constant λ were treated as the parameters to be found.

(ii) EXPFIT (2), which subtracted A_1 (assumed constant and fed in as input data) from the data and fitted the resulting data to the form $A_2 e^{-\lambda t}$ where the parameters of fit were the amplitude A_2 and the decay constant.

Both these routines calculated the variance of fit, defined as

$$\text{VAR} = \sqrt{\sum \frac{w_i \, (y_i - y \, (A_1, A_2, \lambda) \,)^2}{N - 3}} \quad ,$$

where w_i = weight of each point,

y_i = experimental values,

$y \, (A_1, A_2, \lambda)$ = calculated values,

and N = number of points in the fitted curve.

This is both a measure of how good the fit is and how well the weights have been chosen. Both routines were also designed to fit the data from progressively later times after the initial pulse to some specified last channel.

4. RESULTS

4.1 Results of Exponential Fitting

Eighteen assemblies covering the buckling range 1.93×10^{-2} cm^{-2} to 5.34×10^{-2} cm^{-2} were studied, and decay curves for the three different time regions described above were measured for each assembly. In all attempts to fit an exponential, the data from some chosen starting time (channel) to the end of the measured time distribution was used. By using the stepping facility of the routines described above about twenty fits were made to the data in each time region allowing any change of the decay constant with time to be observed. If

the decay curve in a given time region can be described by a single exponential then the fitted decay constant should not change in any systematic way with time.

The decay of the neutron population in the assemblies has been observed from immediately after the pulse and thus at early times one can expect non-exponential behaviour due to thermalisation effects and the existence of non-negligible higher order spatial modes. $^{239}Pu/BF_3$ ratio measurements indicated that neutron thermalisation effects are negligible after about 1.5 ms. This result compares favourably with the 2 ms for graphite suggested by Cuny et al. (1965). On this basis we define an 'exponential criterion' thus: a discrete exponential decay exists if the fitted decay constant remains constant for all times greater than ~ 1.5 ms after the pulse.

The results of fits were plotted in the form of fitted λ versus starting channel (of fit) curves. Figure 4 shows results that indicate exponential, doubtful and non-exponential decays respectively. These diagrams show the time of the start of the fit and the time interval over which the decay was observed.

Initially all results were analysed using EXPFIT (1). This routine, which fits three parameters (A_1, A_2 and λ) had the undesirable effect of fitting both a varying decay constant and a varying background in non-exponential cases even though the background was shown experimentally to be constant. To check that the observed variations in decay constant did in fact indicate non-exponential decay, the data was also analysed using the background constraining EXPFIT (2). In this case a background indicated by the results of EXPFIT (1) and also consistent with the decay curves was used and the fit to two parameters was examined. Figure 5 shows, as an example, the results of this procedure applied to the time region II for the 17.75 x 30.48 x 30.48 cm^3 assembly. The diagram shows the series of fitted decay constants, backgrounds and variances of fit when EXPFIT (1) was used, compared with the series of fitted decay constants and variances of fit when EXPFIT (2) was applied. It is obvious that a non-exponential decay exists in this case.

The composite decay curve of the 17.78 x 30.48 x 30.48 cm^3 assembly has been plotted (Figure 6) for the full time range investigated by normalising the region I, II and III curves in the areas of overlap. The deviation from exponential decay, although slight on this scale, has been emphasised by drawing a straight line in the diagram.

Figure 7 shows the results of investigations on fourteen small assemblies.

This diagram is constructed so that the assembly shape is emphasised. Assemblies which presented the same cross section normal to the incident neutron beam are plotted on a line opposite the corresponding buckling and the type of decay (exponential, doubtful or non-exponential) is indicated. The buckling of each assembly has been calculated by making corrections for voidage (Ritchie 1967), normalising the density to 2.96 g cm^{-3} and using an extrapolation length of 1.149 cm. Even though the meaning of buckling is suspect in the region under consideration it has been retained as a useful parameter which approximates leakage in terms of the assembly dimensions.

4.2 Effect of Different Source and Counter Conditions

The effects of source pulse width, detector position and detector size on the results for non-exponential decay of the neutron population was examined in the 17.78 x 30.48 x 30.48 cm^3 stack where the decay was markedly non-exponential and thus sensitive to changes in these parameters. Figure 8 shows the results of the investigation with:

(i) the normal arrangement with two 1 in. BF_3 detectors at the top centre of the assembly with their lengths parallel to the beam direction and a source pulse width of 500 μs,

(ii) two 1 in. BF_3 detectors at the top centre of the assembly with their lengths parallel to the beam direction and a source pulse width of 250 μs,

(iii) two 1 in. BF_3 detectors on top of the assembly on the centre line of the stack perpendicular to the beam direction but at one edge of the assembly and a source pulse width of 500 μs,

(iv) one 1 in. BF_3 detector at the top centre of the assembly with its length parallel to the beam direction and a source pulse width of 500 μs.

The diagram is drawn as a fitted decay constant versus starting channel curve with results for corresponding starting times side by side and the various sets of fits separated by dotted lines.

The examination of the dependence of the decay on source pulse width indicates a slightly faster decay for the shorter (250 μs) source pulse width. The mean lifetime of the decay in this stack was ~ 200 μs so that the pulse widths used were approximately 1.25 mean lives (250 μs) and 2.5 mean lives (500 μs). Hence the system was in different stages of neutron saturation at the end of these two

different pulses, and any marked dependence of the decay on the source pulse width should be apparent. No large effect is apparent.

There is no significant difference between the results of observation with two detectors in parallel and with a single detector, both at the top centre of the stack and with their lengths in the beam direction. However, the time distribution measured by the two detectors on the top surface of the stack but displaced to one edge indicates a slightly slower rate of decay.

4.3 The $\lambda(B^2)$ Curve

The decay constants of those curves which could be described by an exponential have been normalised to a density of 2.96 g cm^{-3}, corrected for an absorption corresponding to 1.954×10^2 sec^{-1} (Ritchie 1967) and plotted as a function of buckling (also normalised to 2.96 g cm^{-3}) in Figure 9. For those curves that could not be described by an exponential it was found that the 'decay constant' from the fit at progressively later times after the pulse varied in quite a smooth manner throughout the time regions I, II and III (for example Figure 4). For these curves the effective 'decay constants' as given by the fit to an exponential at waiting times of 1.0, 1.5, 2.0 and 2.5 ms after the pulse were also normalised to the reference density, corrected for the absorption and plotted as a function of buckling in Figure 9.

Also shown in Figure 9 are the limits proposed by Corngold and Kothari for the upper bound on the value of the discrete mode of decay. These limits were evaluated using two sets of data: that from the work of Bhandari et al. (1958) and that from cross sections evaluated at this laboratory (Maher and Trimble - A.A.E.C. unpublished data). In the latter case the coherent elastic cross section of beryllium oxide was evaluated exactly from the known crystal structure and the scattering cross sections of beryllium and oxygen. The inelastic scatter was assumed to be well represented by the incoherent inelastic cross section. This was evaluated by subtracting the incoherent elastic cross section, which can be calculated analytically from the total incoherent cross section. This last was produced by the code PIXSE (MacDougall 1963) using an $S(\alpha,\beta)$ from the code LEAP (McLatchie - unpublished) and using Sinclair's $\rho(\beta)$ to represent the phonon frequency distribution in beryllium oxide. In evaluating $(v\Sigma_{inel})_{min}$ it was assumed that the inelastic cross section is proportional to $1/v$ at the Bragg cut-off (0.0037 eV).

Other evaluations of $(v\Sigma_{inel})_{min}$ have been given elsewhere (Ritchie and Rainbow 1967, Trikha 1965) but these are in general somewhat lower than those

given in Figure 9. Zhezherun (1967) has indicated that the grain size of a given polycrystalline sample may well reduce the scattering cross section below the theoretical value. This could make the transport cross section maximise at 0.0157 eV instead of at 0.005 eV. Hence Kothari's expression has also been evaluated at 0.0157 eV where it has also been assumed that the cross section has been reduced by some 20 per cent. This is marked as Kothari's limit II in Figure 9.

5. DISCUSSION

5.1 Comparison with Previous Results

Zhezherun (1967), Joshi et al. (1967) and Iyengar et al. (1957) have studied beryllium oxide assemblies with large bucklings (greater than 3×10^{-2} cm^{-2}) by the pulsed neutron technique. Iyengar et al. do not record non-exponential behaviour even though they examined assemblies with bucklings of up to 6.1×10^{-2} cm^{-2}. This work was done with less sophisticated equipment than is now available before the significance of continuum effects in polycrystalline moderators was realised. The pulsing period was only 1.82 ms and the time analyser had only ten channels, an inadequate system for this study. Points read from the $\lambda(B^2)$ curve of Iyengar et al. have been corrected for absorption and plotted in Figure 9. These results are much lower than the results presented here. Zhezherun did not observe non-exponential behaviour for assemblies with bucklings below the large value of 5.6×10^{-2} cm^{-2} (normalised to a density of 2.96 g cm^{-3}). He claims agreement with Kothari's limit calculated by Trikha with data corresponding to the second major Bragg peak in the transport cross section. Points read from Zhezherun's $\lambda(B^2)$ curve and normalised to a density of 2.96 g cm^{-3} have been plotted in Figure 9. The results of Joshi et al. who observed exponential behaviour up to $B^2 = 5.97 \times 10^{-2}$ cm^{-2}, have also been corrected for density and absorption and plotted in Figure 9.

Zhezherun's results are significantly higher than the present ones throughout the whole buckling range. In particular they are higher than the present ones below $B^2 = B_*^2 = 2.1 \times 10^{-2}$ cm^{-2} where the present ones are in good agreement with those of Cuny et al. (1965) and Joshi et al. (1965). The discrepancy is hard to explain unless Zhezherun's sample of BeO has scattering properties different from those of the other samples, a possibility noted by Zhezherun himself who discusses it and its effect on Kothari's limit in some detail.

The fact that Zhezherun does not observe non-exponential behaviour until the buckling reaches much larger values could be because his counting statistics are

a factor of ten lower than those in the present experiment in the time region of interest. Data from the decay curve of the 17.78 x 30.48 x 30.48 cm^3 assembly covering the same time region investigated by Zhezherun (1.0 to 2.0 ms) produced decay constants which varied by about 1 per cent when the 'waiting' time after the pulse was changed from 915 μs to 1,281 μs. This is similar to the change observed by Zhezherun but the precision of his measurement at a waiting time of 1,120 μs was only 5.5 per cent compared to a precision of 0.3 per cent for the present measurement. This, together with the fact that he did not use waiting times larger than 1,120 μs and so saw no systematic change in decay constant with the greater waiting times might explain why he described these decay curves as exponential. Joshi et al. (1967) used even shorter waiting times (~ 500 μs) than Zhezherun. They do not quote any values for the change in the observed decay constant with waiting time, but they do quote errors of between 0.4 and 1.0 per cent in the decay constants at large buckling. These errors indicate errors of between 3 and 10 per cent at waiting times of 1.5 ms. Hence any non-exponential behaviour would again be masked.

Further examination of Figure 9 also shows how it is possible to obtain a fairly smooth $\lambda(B^2)$ curve well into the continuum. If the decay curves are analysed in the same way each time, for example, by using the same waiting time after the pulse, then the 'decay constants' will vary in a smooth manner with increasing buckling. This is particularly so if the waiting time is short (~ 1.0 ms).

5.2 Discussion of the $\lambda(B^2)$ Curve

The shaded region in Figure 7 defines an experimentally observed transition region between discrete exponential decay of the neutron population in beryllium oxide assemblies and a continuous mode of decay. This allows us to define in terms of buckling an experimental upper bound to discrete modes. Thus

$$(B^2_*)_{exp} = (3.075 \pm 0.125) \times 10^{-2} \text{ cm}^{-2} .$$

The corresponding experimental upper bound on the decay constant of discrete modes is

$$\lambda^* = (3.65 \pm 0.1) \times 10^3 \text{ sec}^{-1} .$$

It is as well to recall at this point the exponential criterion that is used and to observe that the magnitude of the errors (see Figure 4) associated with the

fitted decay constants are typically of the order 1.0 to 0.4 per cent. This implies that a decay which may, in reality, be slightly non-exponential will be considered as exponential if its decay constant varies in a systematic way by less than 0.5 per cent when fitted to an exponential as described above. The decision as to whether an assembly has a discrete decay mode tends to be subjective to this extent.

Figure 7 does not indicate any obvious dependence of $(B^2_*)_{exp}$ on the assembly shape. We would expect, if there is any shape dependence, that 'slabs' (a x 60.96 x 60.96 cm^3, a x 45.72 x 60.96 cm^3 where $a \leq 20.32$) should have a lower $(B^2_*)_{exp}$ than 'cubes' (~ 30.48 x 30.48 x 30.48 cm^3). Because the minimum dimension in 'slabs' is smaller, neutrons with longer mean free paths (sub-Bragg neutrons) will leak from the assembly more readily, and so at an apparently smaller buckling upset the balance between leakage and energy transfer which maintains the asymptotic energy spectrum. Results indicate that any variation in $(B^2_*)_{exp}$ between the slab-like assemblies and the cube-like assemblies is less than 10 per cent.

The $\lambda(B^2)$ curve (Figure 9) shows that Kothari's limit when calculated using the Lucas Heights data corresponding to the first major peak in the transport cross section agrees very well with our results. Corngold's limit is too low and Kothari's limit calculated using data corresponding to the second major Bragg peak in the transport cross section is too high.

In Figure 9 an envelope has been drawn about the 'effective' decay constants of decay curves not described by an exponential. The interpretation of this is that for a particular buckling greater than $(B^2_*)_{exp}$ the decay constant obtained from fitting the decay curve to an exponential lies somewhere in the shaded area if the waiting time is between 1.0 ms and 2.5 ms after the pulse. It gives some idea of the divergence of the decay from exponential for a particular buckling and indicates that the region $B^2 = 3.0 \times 10^{-2}$ cm^{-2}, $\lambda = 3.65 \times 10^3$ sec^{-1} is definitely some sort of transition region. Corngold and Durgun (1967) show, using diffusion theory and a simple model for the scattering kernel which has the properties of a Bragg cut-off, that the distribution function associated with the continuous mode of decay at buckling values just above $(B^2_*)_{exp}$ is a Laplacian with a large narrow peak. This means that the decay will appear almost exponential over quite a long time period. This is not inconsistent with the present observations between $B^2 \sim 2.1 \times 10^{-2}$ cm^{-2} and 2.95×10^{-2} cm^{-2} in view of the remarks at the beginning of the section on the precision with which experimental decay constants can be measured. However, Corngold and Durgun predict that the width

of the distribution function will increase fairly slowly with buckling. This is not consistent with the rather sharp transition at $B^2 \sim 3.0 \times 10^{-2}$ cm^{-2}.

An examination of the results in the shaded area reveals evidence of a dependence of the rate of decay on the assembly shape. The 'decay constants' of slab-like assemblies decrease faster than cube-like assemblies of the same order of buckling (see for example the group 20.32 x 30.48 x 30.48 cm^3, 17.78 x 30.48 x 45.72 cm^3, 15.24 x 45.72 x 60.96 cm^3). This phenomenon is consistent with the argument presented earlier in the discussion of the shape dependence of the upper bound to the discrete modes. Also the fact that the 'decay constant' of the 17.78 x 60.96 x 60.96 cm^3 assembly (labelled as doubtful) decreases faster than that of the 27.94 x 30.48 x 30.48 cm^3 assembly (also labelled as doubtful) which has a slightly lower buckling, suggests that there is possibly some shape dependence in the upper bound of the discrete mode, this dependence being within the bounds indicated earlier.

Corngold and Durgun (1967) in their recent analysis of the pulsed neutron experiment also presented the result that the $\lambda(B^2)$ curve intercepts the line $\lambda = (v\Sigma_{inel})_{min}$ with zero tangent. Measurements undertaken in this region on assemblies with bucklings of 1.931, 1.993, 2.067 and 2.156 cm^{-2} (see Figure 12) show no tendency for the $\lambda(B^2)$ to exhibit the predicted behaviour. This disagreement is hardly surprising since the theoretical result is almost certainly a property of the diffusion approximation. This is not expected to be a very good approximation in these small pulsed assemblies in which some neutrons have mean free paths of the order of the assembly dimensions. The results in the region of $(v\Sigma_{inel})_{min}$ have been presented in more detail in Table 2.

6. CONCLUSION

The main results of the present experiment on the mode of decay in pulsed beryllium oxide assemblies with decay constants in the vicinity of $(v\Sigma_{inel})_{min}$ can be summarised as follows:

1. In assemblies with bucklings less than about 2.1 x 10^{-2} cm^{-2}, which corresponds to a $(v\Sigma_{inel})_{min}$ of 2.6 x 10^3 sec^{-1}, the decay curve can be described by an exponential after some suitable waiting time has been allowed to elapse.

2. In the buckling range 2.1×10^{-2} cm^{-2} $\leq B^2 \leq 2.91 \times 10^{-2}$ cm^{-2} the decay curve is exponential within the limits of the present determination. In other words the decay constant does not change with time

at waiting times of greater than 1.5 ms where the error in the decay constant in all cases was better than 0.5 per cent at about 3.0 ms after the pulse.

3. In assemblies with bucklings greater than or equal to 3.3×10^{-2} cm^{-2} the decay curve cannot be described by an exponential at waiting times of greater than 1.5 ms.

4. Between 2.91×10^{-2} cm^{-2} and 3.3×10^{-2} cm^{-2} there is a twilight region in which no definite statement can be made about the mode of decay.

These results are apparently in disagreement with those of both Zhezherun and Joshi et al. (1967). It seems likely that the poorer statistics of Zhezherun's experiment in the time region of interest led him to describe a decay curve as exponential when in fact it was changing with time by an amount less than his experimental errors. The very short waiting times used by Joshi et al. probably produced the same effect.

The values presented for $(v\Sigma_{inel})_{min}$ in Figure 9 are the largest of the several quoted in the literature. Hence there is apparently a region of buckling certainly greater than that corresponding to B^2_* in which the decay curve is described accurately by an exponential over a reasonable length of time (1.5 to 3.0 ms) after the pulse. Beyond this there is a surprisingly sharp transition to a state where the decay is not described by an exponential. This point is in good agreement with the limit proposed by Kothari estimated at the major Bragg peak in beryllium oxide (0.005 eV). One might be tempted to take this agreement to mean that Kothari's limit is the correct expression for the upper limit on the discrete mode of decay. However, the analysis used in the derivation of this expression assumes some low energy cut-off in the neutron energy distribution. Since there is no physical reason why this lower cut-off should exist and since if the lower cut-off is allowed to drop below a certain level ($\sim 10^5$ eV), then $(v\Sigma_{inel} + vDB^2)_{v = v_o}$ is no longer the minimum value of $(v\Sigma_{inel} + vDB^2)$, this explanation does not seem sufficient.

It would appear then that any theory on the decay of the neutron population in these small pulsed assemblies must explain why the decay appears to be exponential at bucklings greater than B^2_* and why there is such a sharp transition at a buckling corresponding to $(v\Sigma_{inel} + vDB^2)_{v = v_o}$. If such a theory shows that the decay curves for $B^2 > B^2_*$ are not in fact exponential but differ from exponential by only a small amount (let us say they vary by less than 0.5 per cent over the range 1.0 ms to 4.0 ms), then it seems unlikely that the present integral

methods will provide a reasonable test of the theory. Such a test might be better done by measuring time-dependent energy spectra in the region of $(\nu\Sigma_{inel})_{min}$ and it is to be hoped that any theoretical approach to this problem will give some indication of changes in the energy spectra.

7. ACKNOWLEDGEMENTS

We would like to thank Mr. G. Trimble whose assistance in producing and running the various analysis programmes proved invaluable and also the operating staff of the 3 MeV Van de Graaff accelerator at the A.A.E.C. Research Establishment, Lucas Heights, for their assistance during the experiments. One of us, M. T. Rainbow, would like to thank the Australian Institute of Nuclear Science and Engineering for their generous grant to the University of Tasmania in support of this work, and to acknowledge the continued interest of Dr. A. G. Fenton of the University of Tasmania during these experiments.

8. REFERENCES

Albertoni, S. and Montagnini, B. (1965). - Proceedings of I.A.E.A. Conference on Pulsed Neutron Research, Karlsruhe, Vol. 1, p.239.

Andrews, W. M. (1960). - URCL-6083.

Bhandari, R. C. Kothari, L. S. and Singwi, K. S. (1958). - J. Nucl. Energy, 7 : 45.

Bednarz, R. (1965). - Proceedings of I.A.E.A. Conference on Pulsed Neutron Research, Karlsruhe, Vol 1, p.259.

Corngold, N. (1964). - Nucl. Sci. Engng, 19 : 80.

Corngold, N. and Durgun, K. (1967). - Nucl. Sci. Engng, 29 : 354.

de Saussure, G. (1962). - Nucl. Sci. Engng, 12 : 433.

Fraser, H. J., Ritchie, A. I. M. and Whittlestone, S. (1968). - Rev. Sci. Instrum, 39 : 240.

Gaerttner, E. R., Daitch, P. B.,Fullwood, R. R., Lee, R. R. and Slovacek, R. E. (1965). - Proceedings of I.A.E.A. Conference on Pulsed Neutron Research, Karlsruhe, Vol. 1, p.483.

Iyengar, S. B. D., Mani, G. S., Ramanna, R. and Umakanth, N. (1957). - Proceedings of Ind. Acad. of Sci. 45(A) : 215.

Jha, S. S. (1960). - J. Nucl. Energy, 12 : 89.

Joshi, B. V., Nargundkar, V. R. and Subbarao, K. (1965). - Proceedings of I.A.E.A. Conference on Pulsed Neutron Research, Karlsruhe, Vol. 1, p.405.

Joshi, B. V., Nargundkar, V. R. and Subbarao, K. (1967). - J. Nucl. Energy, 21 : 883.

Kothari, L. S. (1965). - Nucl. Sci. Engng, 23 : 402.

Kuscer, I. and Corngold, N. (1965). - Phys. Rev, 139A : 981.

MacDougall, J. K. (1963). - AEEW-M318.

Ritchie, A. I. M. (1967). - AAEC/TM415.

Ritchie, A. I. M. and Rainbow, M. T. (1967). - Nucl. Sci. Engng, 28 : 306.

Silver, E. G. (1962). - Proceedings of Brookhaven Conference on Neutron Thermalisation, BNL-719, Vol. 3, p.981.

Trikha, S. K. (1965). - J. Nucl. Energy, Parts A and B, 19 : 687.

Zhezherun, I. F. (1964). - J. Nucl. Energy, 18 : 279.

Zhezherun, I. F. (1967). - Proceedings of I.A.E.A. Conference on Neutron Thermalisation and Reactor Spectra, Ann Arbor.

TABLE 1

$(v\Sigma)_{min}$ FOR VARIOUS MODERATORS

Moderator	Comments			Density ($g\ cm^{-3}$)	$(v\Sigma)_{min}$ (sec^{-1})
	Gas	A	σ_s (barns)		
Free monatomic gas	proton	1	20.6	4.46×10^{-5}	137.9
$(v\Sigma)_{min} = \left(\frac{8\ kT}{\pi\ mA}\right)^{\frac{1}{2}} \Sigma_s$	'beryllium'	9	6.18	4.02×10^{-4}	13.74
	'carbon'	12	4.64	5.36×10^{-4}	9.07
	'oxygen'	16	3.76	7.14×10^{-4}	6.29
H_2O				1.00	300,000
Beryllium				1.85	3,800
Carbon				1.60	2,600
Beryllium oxide				2.96	2,500

A = mass number of gas

m = mass of neutron

σ_s = free atom scattering cross section

TABLE 2

DECAY CONSTANTS OF STACKS WITH BUCKLINGS IN THE REGION OF $B_x^2 = 2.1 \times 10^{-2}$ cm^{-2}

Nominal Dimensions of Stack (cm^3)	Normalised Buckling (cm^{-2})	Normalised Decay Constant (sec^{-1})	Normalised $\delta\lambda$
43.18 x 30.48 x 45.72	1.931×10^{-2}	2.623×10^3	0.003×10^3
40.64 x 30.48 x 45.72	1.993×10^{-2}	2.696×10^3	0.004×10^3
38.10 x 30.48 x 45.72	2.067×10^{-2}	2.778×10^3	0.004×10^3
35.56 x 30.48 x 45.72	2.156×10^{-2}	2.879×10^3	0.004×10^3

Note: No correction has been made for $\lambda_a = 1.954 \times 10^2$ sec^{-1}

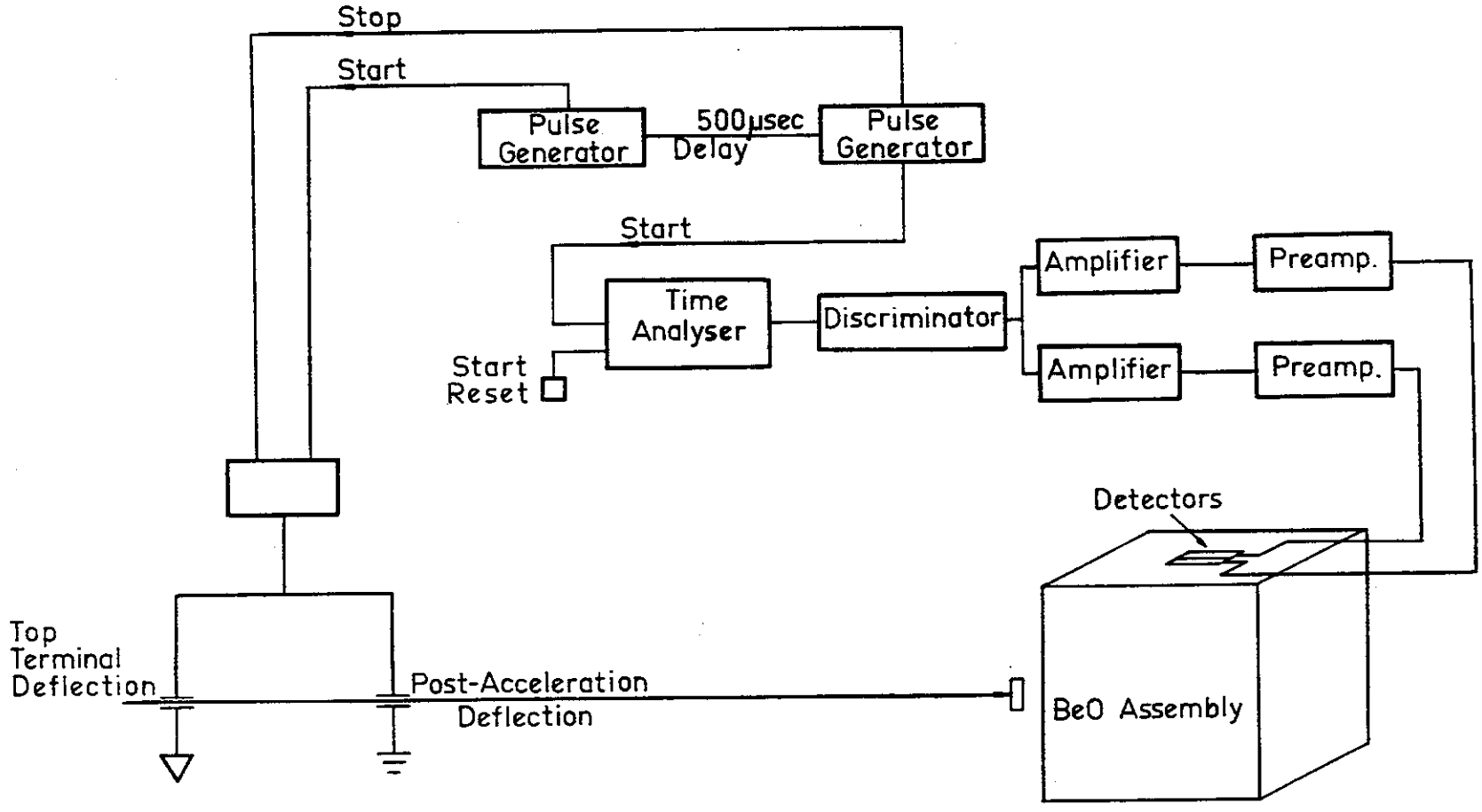

FIGURE 1. ELECTRONICS AND TIMING SYSTEM FOR MEASURING TIME DISTRIBUTIONS IN BERYLLIUM OXIDE

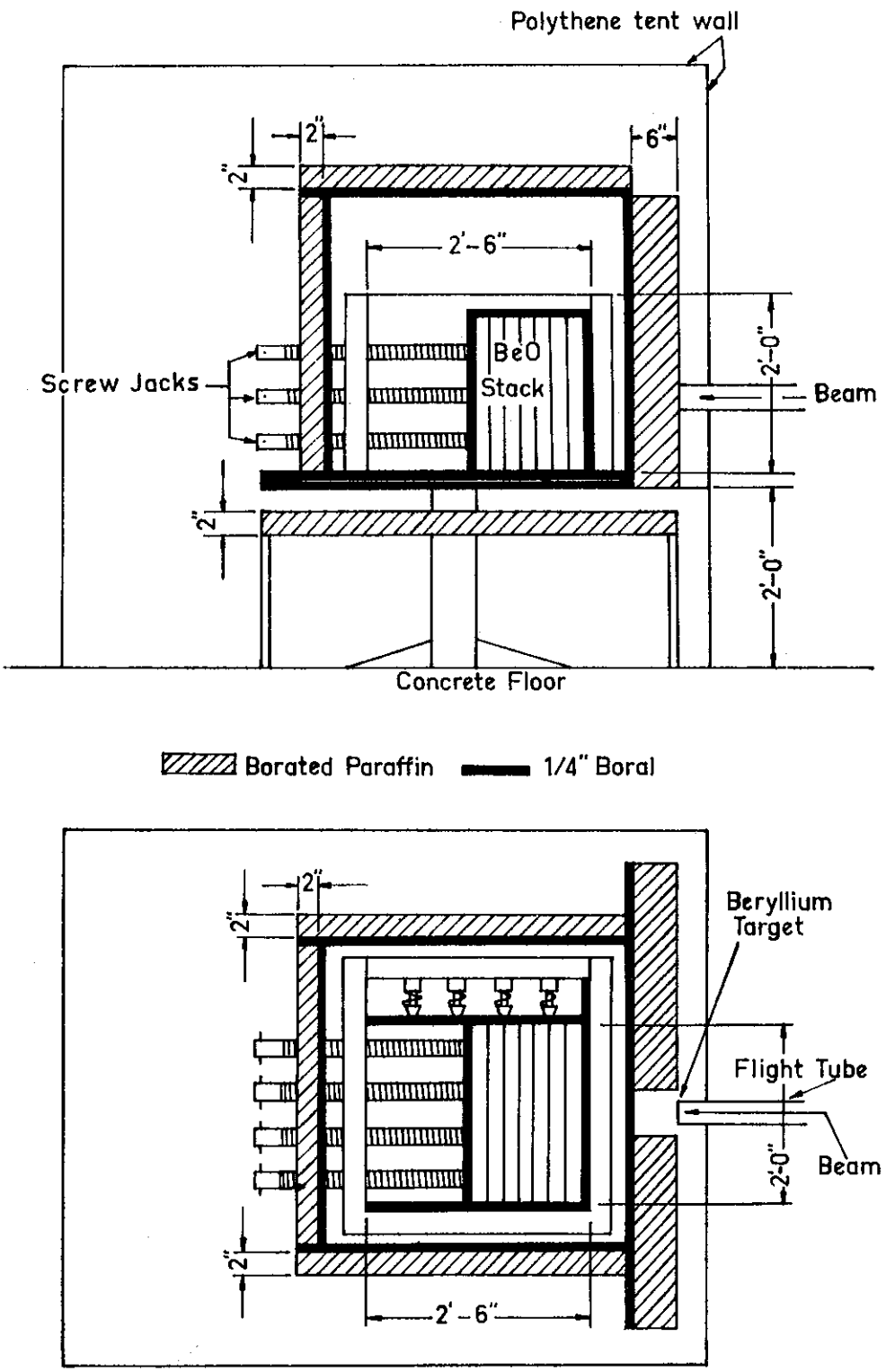

FIGURE 2. SCHEMATIC DIAGRAM OF BERYLLIUM STACK AND ASSOCIATED SHIELDING

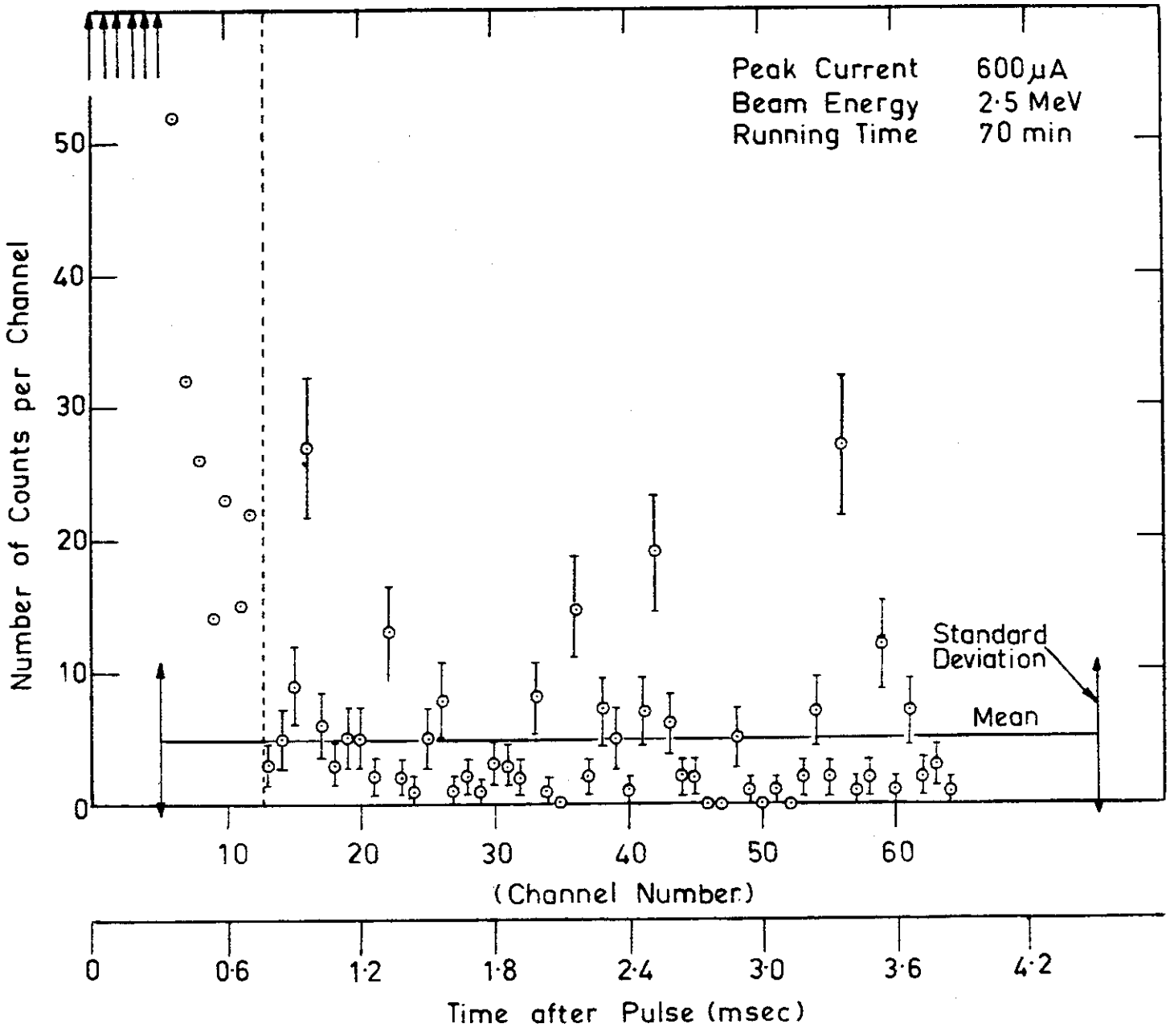

FIGURE 3. TIME DISTRIBUTION OF EMPTY ASSEMBLY

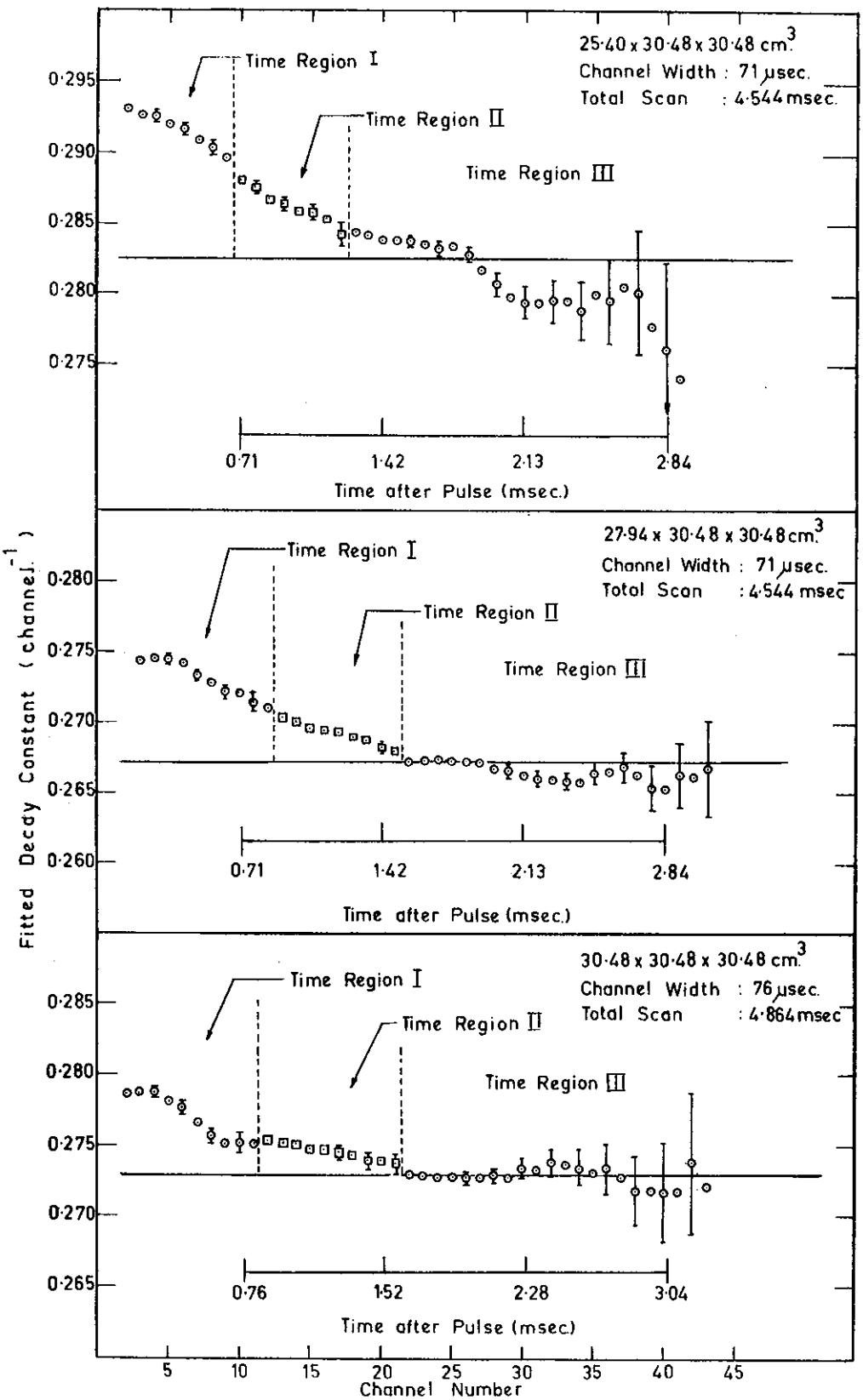

FIGURE 4. ASSEMBLIES EXHIBITING EXPONENTIAL, DOUBTFUL, AND NON-EXPONENTIAL BEHAVIOUR

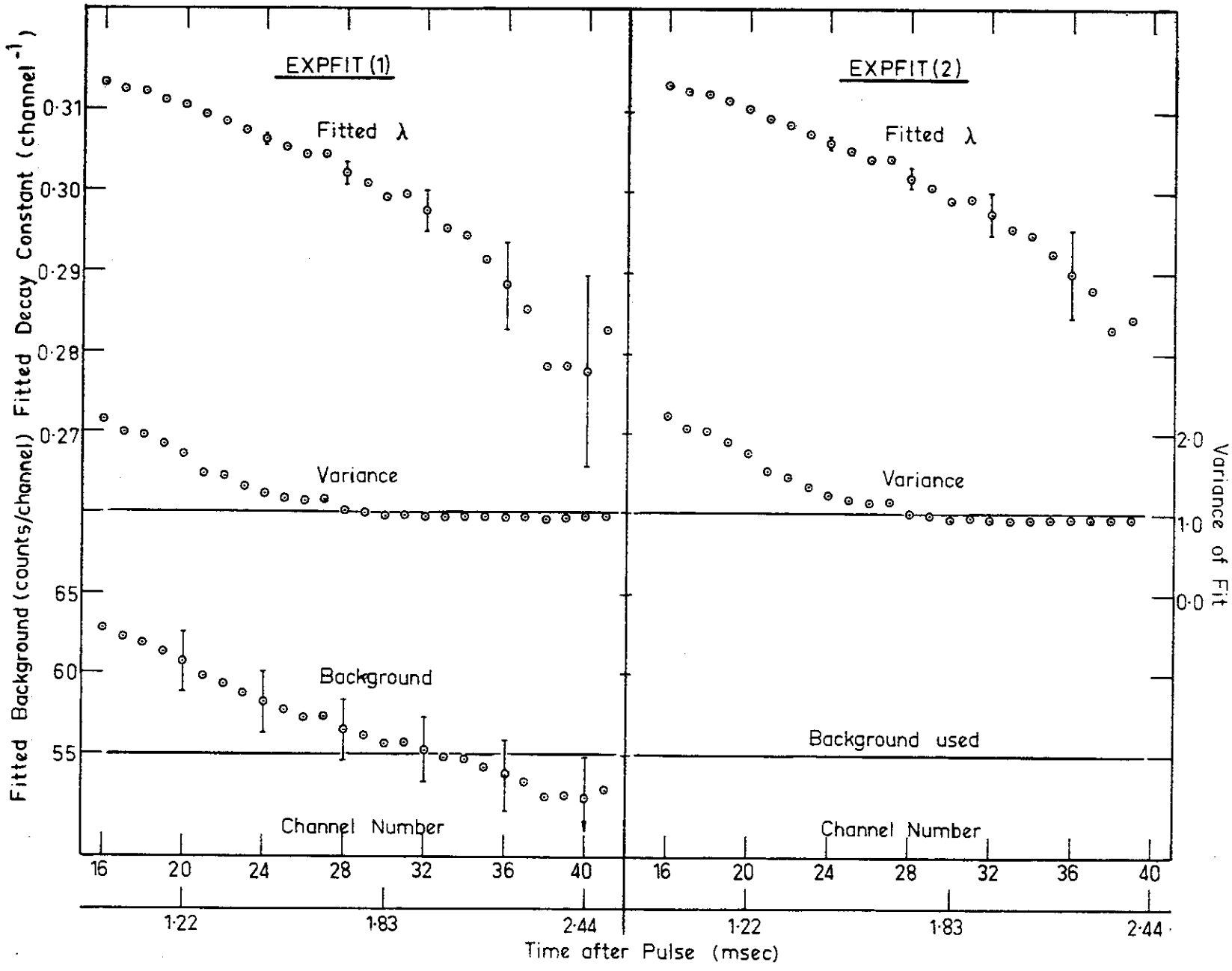

FIGURE 5. ANALYSIS OF 17.78×30.48×30.48 cm^3 ASSEMBLY IN TIME REGION II

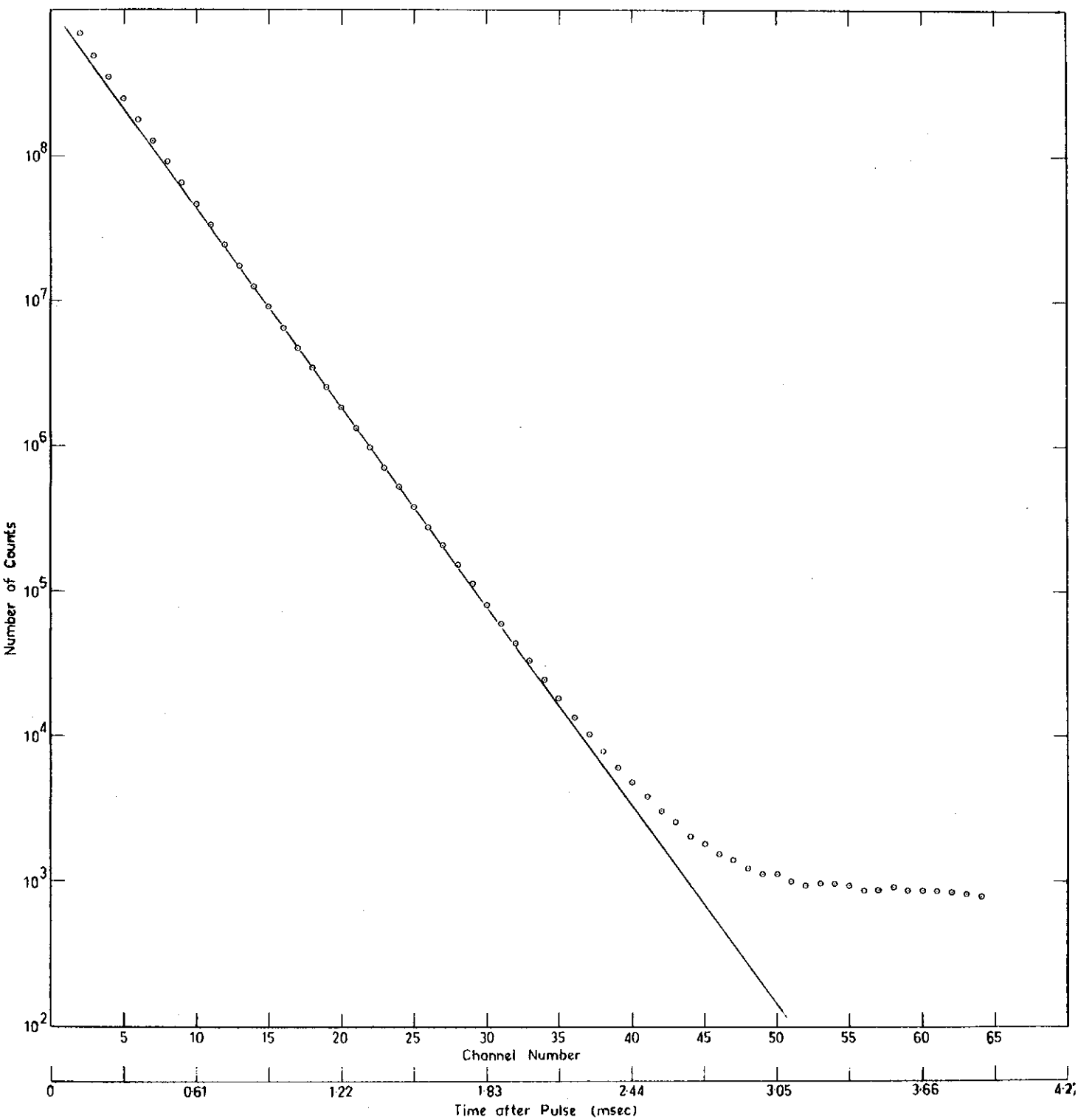

FIGURE 6. COMPOSITE TIME DISTRIBUTION OF 17.78×30.48×30.48 cm^3 ASSEMBLY

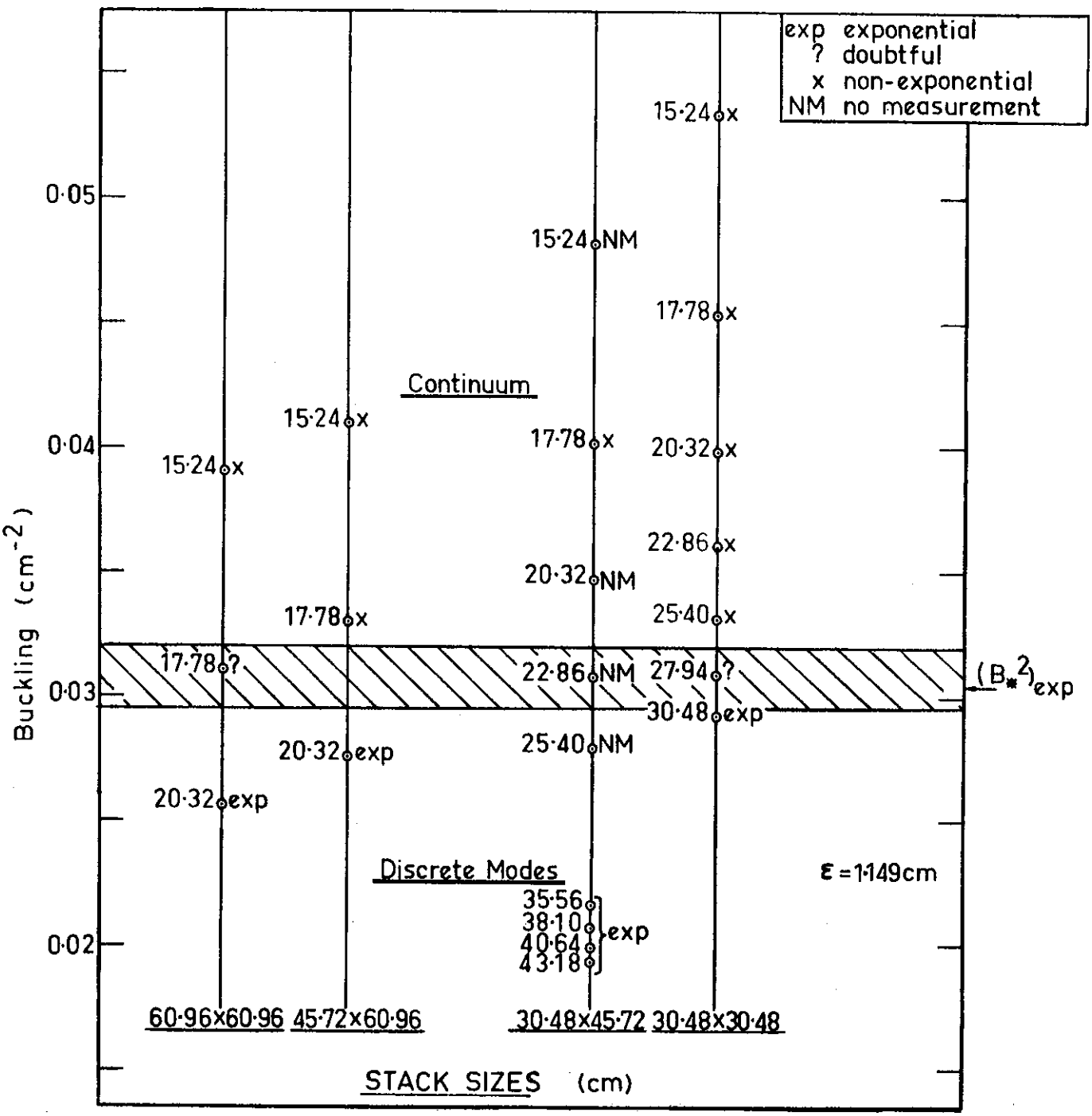

FIGURE 7. DECAYS CONSTANTS NEAR λ^{*}_{exp}

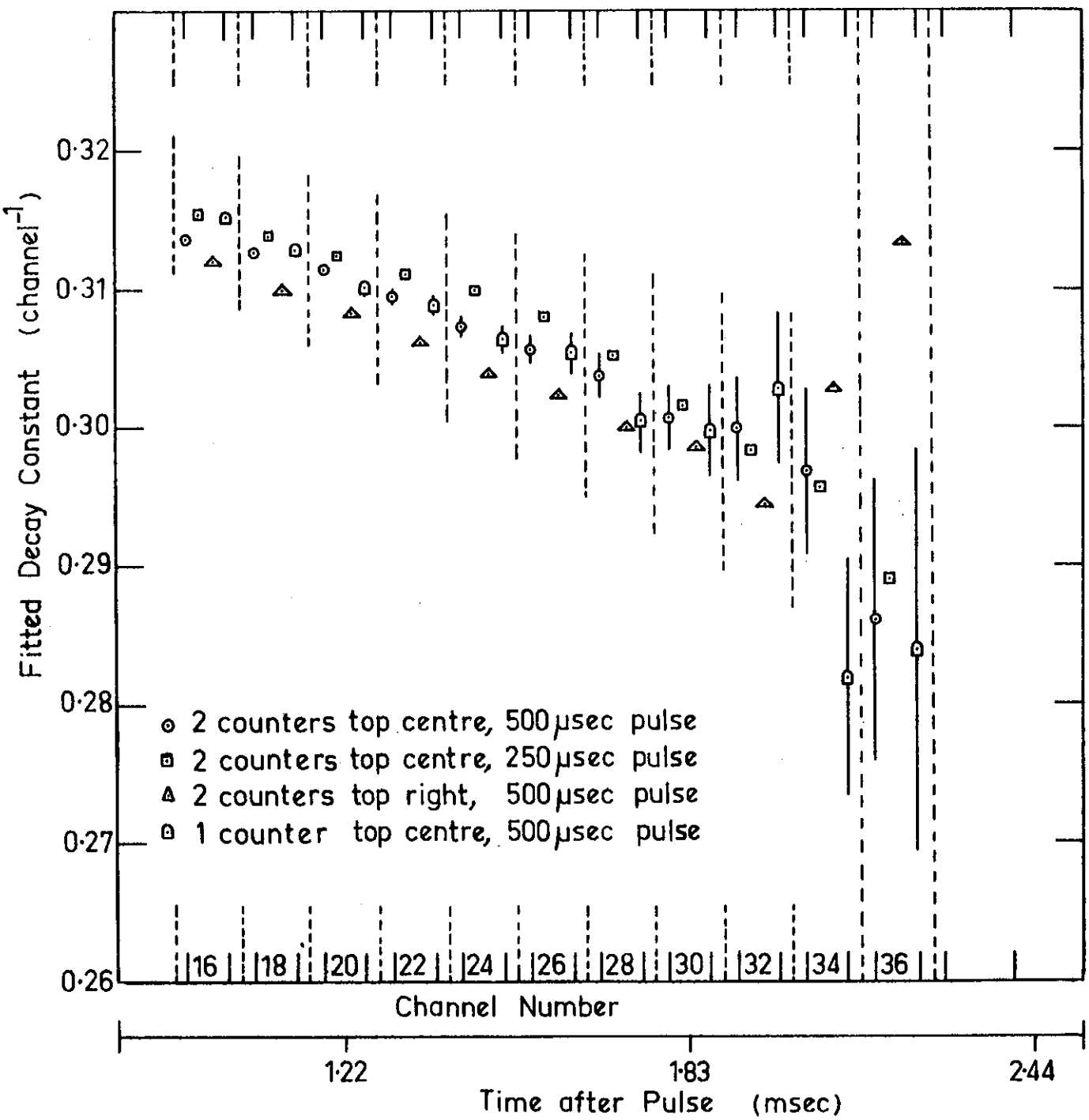

FIGURE 8. INVESTIGATION OF SOURCE WIDTH, DETECTOR SIZE AND POSITION DEPENDENCE OF NEUTRON DECAY USING THE 17.78×30.48×30.48cm^3 ASSEMBLY

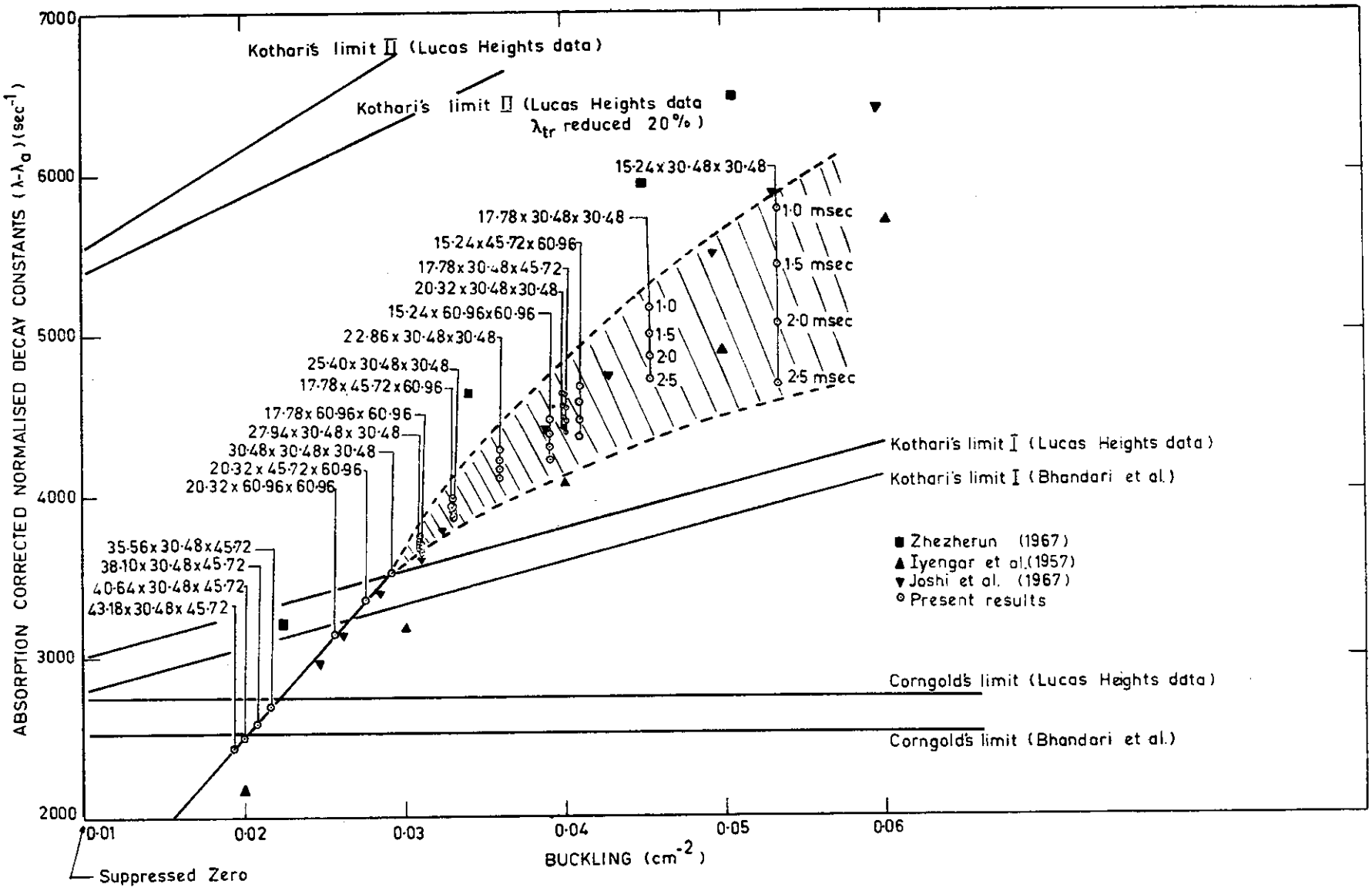

FIGURE 9. NORMALISED $\lambda(B^2)$ CURVE FOR BeO WITH DECAY CONSTANTS IN THE REGION OF λ^*