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FOREWORD

In recent years there has been growing interest in transient, as opposed to static, techniques of neutron and reactor physics investigation. Perhaps the most notable example of the newer transient techniques is the pulsed neutron source method which has been applied to various studies of the detailed interaction of neutrons with non-multiplying as well as multiplying media.

More than 230 scientists from 22 countries and 3 international organizations participated in this IAEA Symposium held at the Kernforschungszentrum Karlsruhe, at the invitation of the Government of the Federal Republic of Germany. Although there have been previous meetings concerned with pulsed neutron measurements (notably at Berkeley in 1958 and at Brookhaven in 1962), this was the first international meeting on pulsed neutron research in which working scientists from all parts of the world participated.

This Symposium has provided not only new and significant pulsed neutron data, but also, what is equally important, fresh viewpoints of interpretation pointing toward useful directions for future pulsed neutron investigation. The general opinion of participants at Karlsruhe was that the potential value of pulsed neutron studies, on fast systems particularly, has scarcely been tapped and that much greater effort in this direction can be expected in the coming years.

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ERRATA

Vol.I, first page of the Contents, referring to the paper printed on page 89. The names of the authors should be as follows:

G. Cuny, V. Deniz, J. Lalande, J.G. Le Ho et M. Sagot (France)

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R. Comte

NON-MULTIPLYING SYSTEMS – EXPERIMENT

(Session I)

A REVIEW OF PULSED NEUTRON EXPERIMENTS ON NON-MULTIPLYING MEDIA

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Abstract — Résumé — Аннотация — Resumen

A REVIEW OF PULSED NEUTRON EXPERIMENTS ON NON-MULTIPLYING MEDIA. For the last twelve years, the technique of pulsed neutron sources has been employed with increasing success for the study of the kinetic behaviour of neutrons in matter. Although the most useful applications of the technique are now in the field of reactivity measurements on multiplying media, there is a continuing interest in its use for studying transient and asymptotic neutron phenomena in non-multiplying assemblies.

A condensed review of this latter type of experiment is given, with particular emphasis on progress since the International Conference on Neutron Thermalization which was held in 1962 at the Brookhaven National Laboratory. This review includes a detailed discussion of the experimental condition for the observation of clean "transient" and "asymptotic" modes. Results for diffusion parameters are summarized and, wherever possible, compared with calculations based on experimental values of the scattering law.

Some new applications of the technique are discussed, for example :

(1) Friedmann's method for the determination of thermalization parameters;

(2) The simultaneous observation of space and time transients in pulsed moderators;

(3) Fast neutron field decay in non-multiplying assemblies of heavy materials.

Finally, a critical comparison between the pulsed source technique and other methods for the investigation of transient and asymptotic neutron phenomena (static methods; harmonically modulated sources) is made.

APERÇU D'ENSEMBLE DES EXPÉRIENCES SUR DES MILIEUX NON MULTIPLICATEURS AU MOYEN DES NEUTRONS PULSÉS. Depuis une douzaine d'années, on utilise avec un succès croissant la méthode des sources de neutrons pulsés pour l'étude du comportement cinétique des neutrons dans la matière. Les applications les plus utiles de cette méthode portent actuellement sur les mesures de la réactivité de milieux multiplicateurs, mais on s'intéresse toujours à son emploi pour l'étude des phénomènes neutroniques transitoires et asymptotiques dans des assemblages non multiplicateurs.

L'auteur donne un aperçu d'ensemble des expériences de ce deuxième type en insistant surtout sur les progrès accomplis depuis la Conférence internationale sur la thermalisation des neutrons, tenue à Brookhaven en 1962. Il discute en détail la condition expérimentale nécessaire pour l'observation des modes « transitoire » et « asymptotique » sans poison. Il fait la synthèse des résultats concernant les paramètres de diffusion et, chaque fois que c'est possible, les compare aux résultats des calculs fondés sur les valeurs expérimentales de la loi de diffusion.

Il décrit certaines applications nouvelles de la méthode et notamment:

1. La méthode de Friedmann pour la détermination des paramètres de thermalisation.

2. L'absorption simultanée des états transitoires variables dans l'espace et dans le temps, dans les ralentisseurs pulsés.

3. La décroissance du champ des neutrons rapides dans des assemblages non multiplicateurs en matières lourdes.

Il procède enfin à une comparaison critique de la méthode de la source pulsée avec d'autres méthodes utilisées pour l'étude des états transitoires et asymptotiques des neutrons (méthodes statiques; sources à modulation harmonique).

ОБЗОР ЭКСПЕРИМЕНТОВ С ИМПУЛЬСНЫМИ НЕЙТРОНАМИ В НЕРАЗМНОЖАЮЩЕЙ СРЕДЕ. В последние двенадцать лет все более успешно используется метод источников импульсных нейтронов для изучения кинетического поведения нейтронов в веществе. Хотя наиболее успешно этот метод применяется теперь в области измерений реактивности в размножающей среде, проявляется постоянный интерес к его использованию при изучении переходных и асимптотических нейтронных явлений в неразмножающих сборках.

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Дается подробный обзор этого последнего типа экспериментов, особо отмечаются успехи, достигнутые со времени Международной конференции по термализации нейтронов в Брукхейвене в 1962 году. Обзор включает детальное обсуждение экспериментальных условий для наблюдения чистых "переходных" и "асимптотических" форм. Результаты по диффузионным параметрам суммируются и, когда это возможно, сравниваются с расчетами, основанными на экспериментальных значениях закона рассеяния.

Обсуждаются некоторые новые методы, например:

1. Метод Фридмана для определения параметров термализации,
2. Метод одновременного наблюдения переходных характеристик во времени и пространстве при импульсе в замедлителе.
3. Метод спада поля быстрых нейтронов в неразмножающих сборках из тяжелых материалов.

В заключение критически сравнивается метод импульсных источников с другими методами исследования переходных и асимптотических нейтронных явлений (статические методы, гармонически модулированные источники).

ESTUDIO PANORAMICO DE DIVERSOS EXPERIMENTOS CON NEUTRONES PULSADOS EN MEDIOS NO MULTIPLICADORES. En el curso de los últimos doce años la técnica de los neutrones pulsados se ha venido empleando con éxito creciente para estudiar el comportamiento cinético de los neutrones en la materia. Aunque las aplicaciones más útiles de esta técnica corresponden actualmente a la esfera de las mediciones de la reactividad realizadas en medios multiplicadores, subsiste el interés por su empleo en el estudio de los fenómenos neutrónicos transitorios y asintóticos en conjuntos no multiplicadores.

El autor ofrece un estudio resumido de esta última clase de experimentos, atendiendo especialmente a los progresos conseguidos desde 1962, año en que se celebró en Brookhaven la Conferencia Internacional sobre Termalización Neutrónica. El estudio panorámico incluye un examen detallado de las condiciones experimentales necesarias para la observación de los modos «transitorios» y «asintóticos», sin deformaciones. En él se resumen, además, los resultados obtenidos para los parametros de difusión, y se comparan - cuando es posible - con los de cálculos realizados partiendo de valores experimentales obtenidos con arreglo a la ley de dispersión.

El autor examina algunas de las nuevas aplicaciones de la mencionada técnica, entre ellas las siguientes:

1. Determinación de los parámetros de termalización por el método de Friedmann.
2. Observación simultánea de los fenómenos transitorios de carácter espacial y temporal en moderadores sometidos a los efectos de los neutrones pulsados.
3. Decrecimiento del campo de neutrones rápidos en conjuntos no multiplicadores de materiales pesados.

Por último, el autor establece una comparación entre la técnica de los neutrones pulsados y otros métodos de investigación de los fenómenos neutrónicos transitorios y asintóticos (métodos estáticos, fuentes armónicamente moduladas).

1. INTRODUCTION

For the last twelve years, there has been an increasing interest in the use of pulsed source techniques for the study of neutron migration, thermalization and absorption in matter. For instance, about twenty different determinations of the diffusion parameters of ordinary water and ice, more than thirty experiments on various "organic" moderators - many of them as a function of temperature - and nine experiments on graphite have been published. Much of the data determined in this way is in disagreement and it appears doubtful if its use in reactor design has justified this enormous effort. There is no doubt, however, that these studies have, probably more than any other class of experiments, contributed to a better understanding of the kinetic behaviour of neutrons in matter. In particular, they have stimulated important developments in transport and thermalization theory, some of which are reviewed at this Symposium [1]. Furthermore, neutron kinetic studies

with pulsed sources have proved to be a useful and rather inexpensive field for student training and in fact a growing percentage of the work is being carried out at university laboratories. Finally, the good success which the pulsed source technique is having for reactivity determinations on reactor systems would probably not have been encountered without the important developments in instrumentation and analysis techniques achieved during the early studies on non-multiplying media.

In this paper some recent developments in this field are summarized, especially the progress made since the last major international conference which was held at Brookhaven in 1962 [2]. Section 2 deals mainly with decay measurements on thermalized neutron fields, to which by far the greatest research activity has been devoted. In section 3 transient phenomena in moderators, i.e. experiments to measure slowing-down and thermalization times, are considered. Section 4 deals with the fairly new field of "quasi-asymptotic" decay of monoenergetic neutron fluxes in heavy scattering substances.

This is not a reporter-type conference, and a considerable number of individual contributions are to be presented. In order not to anticipate too much the following papers we restrict ourselves to areas which are not otherwise covered. The paper is therefore far from a complete survey.

2. DECAY OF A THERMALIZED NEUTRON FIELD

Under certain limitations, which are now fairly well understood theoretically [3], a thermalized neutron field in a moderator is in a true asymptotic state and decays strictly exponentially with time. The classical approach is to measure the time constant α of this decay and to correlate it with the geometrical buckling B^2 of the assembly. The resulting α versus B^2 curve is analysed in terms of the diffusion parameters of the moderating material. Instead of varying the size of the system, one also can vary the concentration N of an added non-1/v absorber; analysing the resulting α versus N curve one observes parameters characteristic of the thermalization properties of the scattering medium. There are closely related stationary techniques, i.e. measurement of the diffusion length as a function of concentration of a 1/v or non-1/v absorber. The α versus B^2 method is hampered by three basic difficulties: The first is a purely experimental one, viz. the precise determination of α in the presence of non-asymptotic neutrons and background. The second problem is a theoretical one and consists in the calculation of the geometrical buckling from the given dimensions of the scattering medium. Thirdly, there is the problem of analysing the α versus B^2 curve properly. The difficulty to define the geometrical buckling does not necessarily arise in α versus N or stationary poisoning experiments, since they can be performed on nearly infinite media. The other two problems, however, arise in both latter techniques in an analogous form.

2.1. Problems arising in α-determinations

An asymptotic spectrum is rapidly established in hydrogenous moderators, because of their extremely good thermalization properties. "Waiting

time" problems thus do not seriously affect α-measurements if care is taken to eliminate higher spatial modes. Room-return background can be annoying but the assemblies can be easily shielded, since they tend to be quite small. Another source of background, as noted by SILVER [4], may be photoneutrons from the decay of 7.35-s N^{16} produced by the $O^{16}(n, p)N^{16}$ reaction on the oxygen present in water. This reaction has a threshold at 9.6 MeV and will occur if the neutron source employs the $H^3(d, n)He^4$ reaction. Other authors did not find this background troublesome; it can be eliminated completely by using a $H^2(d, n)He^3$ or other low-energy neutron source. The advantage of using lower energy sources is that shielding against room-return neutrons becomes simpler, and it is for this reason that OGRZEWALSKI et al. [5] used a fast chopper at a research reactor as a neutron source. It should be kept in mind, however, that the elimination of higher spatial modes will be the more difficult the lower the energy of the neutron source.

The situation is quite different in crystalline moderators, such as beryllium and graphite, where the trapping of low energy neutrons creates a serious limit beyond which the decay constant cannot grow. This limiting value, $\lim_{v \to 0} v\Sigma_t$, is about 2600 s^{-1} in graphite and 3800 s^{-1} in beryllium; the critical values of the buckling are about 15×10^{-3} cm^{-2} and 40×10^{-3} cm^{-2}, respectively. The above figures for the $\lim_{v \to 0} v\Sigma_t$ are based on very old measurements of the scattering cross-section below the Bragg cut-off and may therefore be greatly in error [6]. Nevertheless, it is obvious that at very high bucklings no clean asymptotic mode will exist. This is borne out in Fig. 1 where the decay of the neutron density in a beryllium block of $B^2 = 73 \times 10^{-3}$ cm^{-2} is plotted according to FULLWOOD, SLOVACEK and GAERTTNER [7]. The decay is nearly exponential at times between 300 and 700 μs after the pulse injection but tends to be slower later on. To demonstrate that the latter effect cannot be attributed to improper background subtraction, these authors measured, in the same set-up, the neutron decay in a polyethylene assembly whose size was chosen to give approximately the same decay rate. This decay rate was purely exponential as may be seen in Fig. 1. A similar experiment on a small graphite stack was performed by KÜCHLE [8]. He found a similar deviation from a purely exponential decay but the statistical accuracy of the data was not sufficient to support any far-reaching conclusion.

Being aware of these difficulties, most recent experimentalists working on crystalline media have confined their measurements to the low B^2 range. They have also established criteria to ensure that the observed decay corresponds to a true asymptotic state. Other groups experimenting on graphite [9-12] have determined the waiting time which must elapse after the injection of the neutron pulse before beginning the evaluation of the decay curve for α. Figure 2 shows the waiting time required to obtain good exponential decay in graphite according to SERDULA [12].

Recently, there has been some conflicting information. DAVIS, DE JUREN and REIER [13] have used a paraffin shield around their graphite stacks, in addition to the usual cadmium lining. Furthermore, their experimental area was shielded by walls of cans filled with borated water. Using a $H^2(d, n)He^3$ neutron source and collimating these neutrons directly on their graphite assemblies to avoid direct leakage of source neutrons into the experimental

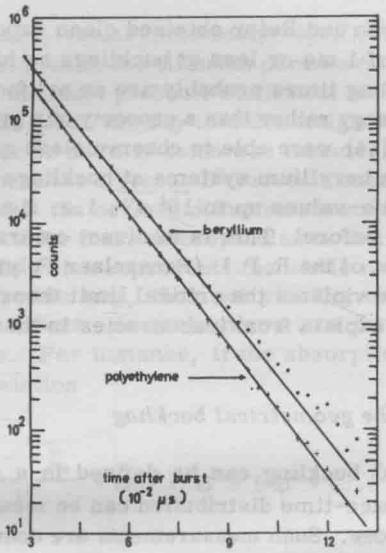

Fig. 1

Neutron die-away in beryllium and in a test assembly of polyethylene selected to give approximately the same decay time (from [7])

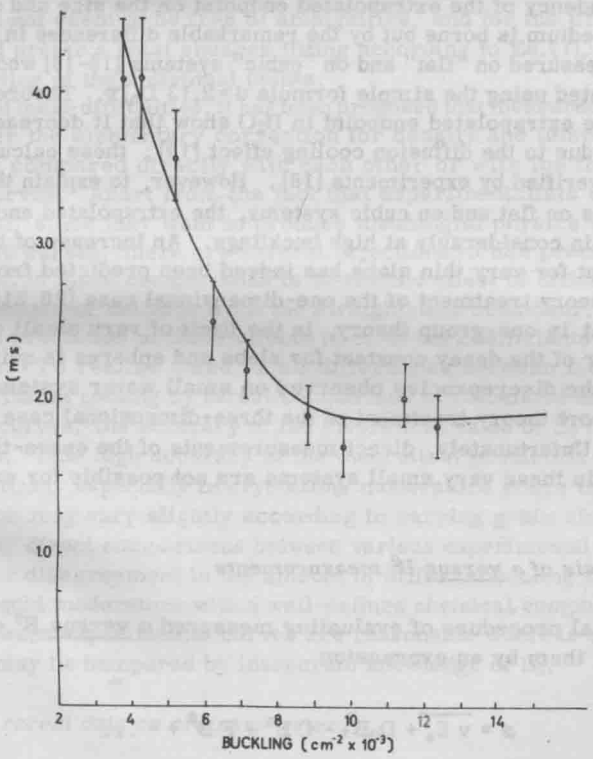

Fig. 2

Time after neutron pulse required for the attainment of an asymptotic spectrum as a function of buckling (from [12])

area, Davis, De Juren and Reier obtained clean exponential decay curves after waiting times of 1 ms or less at bucklings as high as 10×10^{-3} cm^{-2}. They state that "waiting times probably are an artifact of the shielding and the initial neutron energy rather than a property of a graphite stack in space". ZHEZHERUN et al. [14] were able to observe clean exponential decays over up to four decades in beryllium systems at bucklings up to 110×10^{-3} cm^{-2}. This corresponds to α-values up to 10^4 s^{-1}, i.e. figures highly above the critical limit stated before! This is in direct contradiction to the above-mentioned experiment of the R.P.I. (Rensselaer Polytechnic Institute, Troy, N.Y.) group and also violates the critical limit theorem to an extent which would be difficult to explain from inaccuracies in the measured low energy cross-section.

2.2. Definition of the geometrical buckling

The geometrical buckling can be defined in a most straightforward manner if the full space-time distribution can be measured and analysed in terms of Fourier modes. Such measurements are conveniently performed on rather large systems. Experiments on graphite [12, 13] have proved that the relation $d = 2.13\ D_0/\overline{\nu}$ is a very good approximation for the extrapolated endpoint, irrespective of the buckling. For very small water systems, a strong dependency of the extrapolated endpoint on the size and shape of the scattering medium is borne out by the remarkable differences in the α versus B^2 curves measured on "flat" and on "cubic" systems [15-18] whose bucklings were calculated using the simple formula $d = 2.13\ D_0/\overline{\nu}$. Theoretical calculations of the extrapolated endpoint in H_2O show that it decreases with increasing B^2 due to the diffusion cooling effect [19]; these calculations have partly been verified by experiments [18]. However, to explain the measured discrepancies on flat and on cubic systems, the extrapolated endpoint had to increase again considerably at high bucklings. An increase of the extrapolated endpoint for very thin slabs has indeed been predicted from an exact one-group theory treatment of the one-dimensional case [20, 21]. Also, it is known that in one-group theory, in the limit of very small dimensions the behaviour of the decay constant for slabs and spheres is quite different. To resolve the discrepancies observed on small water systems, a multi-group transport theory treatment of the three-dimensional case would be of great help. Unfortunately, direct measurements of the space-time neutron distribution in these very small systems are not possible for experimental reasons.

2.3. Analysis of α versus B^2 measurements

The usual procedure of evaluating measured α versus B^2 curves is to approximate them by an expression

$$\alpha = \overline{\nu \Sigma_a} + D_0 B^2 - C B^4 + F B^6 + - \ldots \qquad (1)$$

using a method of least squares fitting. Some authors use statistical weight

factors on the individual α_i-values (preferably $\sim 1/\alpha_i^2$); others do not. It has been noted that the variation in the diffusion parameters derived in this way may be several times the most probable statistical error, according to the length of the B^2 interval used, the way in which weight factors were applied, or according to whether or not a B^6 form was included. This is especially the case in graphite where the α versus B^2 curves measured by some authors were in agreement whilst the derived diffusion parameters were not. The most popular explanation for these discrepancies is that the above procedure results in a "global" fit which represents the whole measured α versus B^2 region in the best possible way, thus not yielding necessarily the most probable diffusion parameters. Several authors have therefore used different evaluation procedures. For instance, if the absorption cross-section is known, the simpler relation

$$\frac{\alpha - \overline{v\,\Sigma_a}}{B^2} = D_0 - C\,B^2 + F\,B^4 + - \ldots \qquad (2)$$

is fitted to the corrected data. Other authors use iterative procedures, where the weight given to an experimental point is dependent on the influence which the diffusion parameter to be determined has on α. Most of these procedures, however, do not seem to be free of ambiguities, and for the time being the author would prefer a least squares fitting according to Eq. (1), with appropriate weighting of the individual points.

In view of these difficulties, it has been proposed that measured α versus B^2 curves, after the elementary correction for density and temperature deviations, be compared directly with each other or with the theoretically predicted curves. Apart from the fact that experimentalists will not like this proposal, since they want to produce meaningful physics data instead of unanalysed curves, there are several objections to this procedure. Our main interest in these comparisons is to see the effect of diffusion cooling, i.e. the deviation of the data from the straight-line behaviour. We aim at a theoretical prediction of these effects (i.e. of the coefficients C and F) to, say, 15-25%. To realize these small differences between measured and predicted diffusion cooling by direct comparison of calculated and measured α versus B^2 curves, the accuracy of the theoretical value of D_0 must be very high, say 1%. This high accuracy in the theoretical prediction of D_0 seems difficult to attain, especially in crystalline moderators where the scattering cross-section may vary slightly according to varying grain size. For the latter reason, direct comparisons between various experimental curves may show a larger disagreement in the amount of diffusion cooling than actually exists. In liquid moderators with a well-defined chemical composition, comparisons between experimental curves are reasonable whereas comparisons with theory may be hampered by inaccurate knowledge of D_0.

2.4. *Some recent data on ordinary water*

Some results of previous experiments are plotted in Fig. 3. Shown at $B^2 > 0$ are the results of pulsed experiments by KUCHLE [15]. At

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