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Studies of the Fission Integrals of U235

and Pu239 with Cadmium and

Boron Filters

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STUDIES OF THE FISSION INTEGRALS OF U235 AND Pu239 WITH CADMIUM AND BORON FILTERS

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Abstracts

The resonance fissions in U235 and Pu239 have been studied using cadmium and boron filters. Fission chambers were used as detectors and the experiments were performed in beam geometry. The neutron energy distribution in the beams transmitted through the different filters was determined with a fast chopper.

From the cadmium filter measurements the fission resonance integrals were determined. The values obtained were 278 ± 9 b for U235 and 301 ± 10 b for Pu239; 0.5 eV $< E < 1$ MeV.

Complementary Pu239 measurements were made in which the fission events were detected from the fission product activity in irradiated foils. Contrary to what has been reported elsewhere the value of the Pu239 resonance integral, found in this way, agreed well with that obtained from the fission chamber measurement.

The experiments with the boron filters yielded results which, for the thin filter, agreed well with those calculated from the cross section data given in the Karlsruhe compilation. The discrepancy was larger for the thick filter but the values did not disagree outside the common limits of error.

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I.   Introduction

The resonance fissions in U235 and Pu239 above say 0.5 eV play a rather insignificant role for the neutron balance in a thermal reactor system. Even in assemblies with quite hard neutron spectra the percentage of the fissions occurring above 0.5 eV is low, being about 5 % in a spectrum characterized by an r-value (Westcott nomenclature [1]) of 0.1. High accuracy for the fission integrals is therefore not necessarily required for reactor calculations. The values already published in the literature for U235 are well enough in agreement [2, 3, 4, 5[x]] not to warrant any further measurements. For Pu239, on the other hand, the quoted values scatter considerably (op. cit.) and a complementary measurement could be useful. As small fission chambers were constructed at our laboratory for use in a fast assembly, it was thought opportune to include some fission integral measurements during the calibration of the detectors in a thermal reactor. Measurements were made both on U235 and Pu239.

By far the largest contribution to the resonance integral for the two isotopes comes from the low energy region. For Pu239 only about 15 % of the resonance fissions in a $1/E$ flux occurs above 100 eV and the corresponding figure for U235 lies between 20 and 25 %. Calculated values of the fission resonance integral are therefore not very sensitive to the assumed values for the fission cross section in the keV region and upwards. This implies that very little information about these values can be gained by comparing calculated and measured integrals.

To make possible also a check of the high energy cross sections, some additional experiments were made in which boron filters were used to cut out the low energy part of the spectrum. For full use of the results obtained in such experiments, the transmission characteristics of the filters must be known with good precision. These may be calculated if the necessary information concerning mass of boron per unit surface, grain size, enrichment etc. is available. Such data are difficult to obtain accurately. A direct experimental study of the transmitted beam is therefore to be preferred. This has been done in the present investigation.

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[x] The work described in ref. 5 was not published when the present measurements were started.

Cross section information from experiments of such an integral nature as those described in the present report can certainly not be a substitute for direct cross section measurements. However, considering the limited amount of equipment and time required for performing the experiments they have still been considered worth while.

II. Method of measurement

The energy distribution of the neutron flux emerging from the central channel of the Swedish R1 reactor is well-known over a wide energy region from fast chopper measurements. The existence of the chopper also made possible an experimental determination of the particular characteristics of the beams transmitted through the different filters used. The present experiments were therefore performed on the top of the R1 reactor in the beam from the central channel.

The technique employed in the measurements is in many respects similar to that earlier used by Jirlow and Johansson [6] . The relation between the epithermal and thermal flux in the beam is determined from the cadmium ratio of a suitable cross section standard (gold in the present case). Then, assuming the thermal fission cross section to be known for the substance under study, an effective cross section in the epithermal energy region is obtained from the ratio of fissions in the unfiltered and filtered beams. As is well known, measurements with cadmium filters yield for thin samples the dilute resonance integral. With boron filters substituted for the cadmium ones the quantity obtained is less well defined, as will be discussed in some detail later on.

It is customary to include the $1/v$ part in the fission integrals. This custom has been followed here.

III. Experimental procedure

Small fission chambers were used to detect the fission events rather than the fission product activity in irradiated foils. Both methods have earlier been used but in the latter case a source of systematic error is introduced due to variation in the yield figures for the fission products with the energy of the neutrons causing the fissions. Canadian measurements [5] have indicated that this effect could be especially important for Pu239. To get an independent check on this as complementary study on Pu239 with foil technique was made.

The dimensions of the chambers are given in Fig. 1. The coating thickness was 140 $\mu g/cm^2$ for the plutonium chamber and 79 $\mu g/cm^2$ for the uranium one, both layers being thin enough to give negligible shielding corrections. The isotopic concentration for the two materials was as follows (atom per cent).

U235 : 0.95 U234, 93.3 U235, 0.41 U236, 5.34 U238
Pu239: 95.2 Pu239, 4.53 Pu240, 0.25 Pu241.

The corrections to the measured pulse rates because of the presence of other isotopes than Pu239 and U235 were negligibly small.

The chamber under study was placed in a well collimated neutron beam originating from a graphite scatterer placed at the position of maximum flux at the centre of the central channel of the reactor. The arrangement is shown in Fig. 2. The filters were positioned directly on top of the collimator. To avoid the contribution to the pulse rate from neutrons scattered but not absorbed in the filters the detectors were placed at some distance from the collimator. This procedure was considered necessary especially for the thick boron filters for which scattering events reduced the beam intensity by about 15 %.

The measured pulse rates for the boron filter runs were quite small (2-6 p/s). It was therefore essential to keep the contributions from background neutrons low. To achieve this, excess material was avoided in the immediate proximity of the detector, thus reducing the possibility for the neutrons leaking out from the collimator to be scattered in the direction of the chamber. The effect of room scattered neutrons was made small by surrounding the detector equipment at some distance with a boron paraffin shield.

The background was measured with the thick boron filter in place and with a thin paraffin block between the filter and the detector. The paraffin stopped the neutrons in the direct beam from hitting the detector. The counting rate measured was only about 1 % of that obtained without the paraffin block. Thus, background effects were small and could be corrected for.

The cadmium filters were either 1.05 mm or 2.1 mm thick. The epicadmium absorption in Pu239 is quite sensitive to the cadmium thickness, especially for small thicknesses because of the strong 0.3 eV

resonance. Therefore, due to the uncertainty in the transmission function of the filters the value of the plutonium resonance integral deduced from the measurements with the thick filter is more reliable than that obtained with the thin one.

The boron filters consisted of thin boron powder enriched to 90 atom per cent in boron 10. The powder was carefully compressed into aluminium boxes of parallel-epipedic form. Two filters were made, one containing about 0.31 g/cm^2 and the other 0.75 g/cm^2. As the neutron transmission properties of the filters were measured no accurate knowledge of the amount of boron 10 per unit surface was required.

The gold foils used for the calibration were made of a lead-gold alloy, 0.1 mm thick containing 0.1 % by weight of gold. The size of the foils was 4 mm x 30 mm, i.e. somewhat larger than that of the coated chamber electrode (compare Fig. 1). The neutron beam from the collimator was quite narrow and it was important to position the foils accurately. To achieve this the beam was scanned with the chamber to find the best position for the foil holder. The beam profiles determined are shown in Fig. 3a and b.

The neutron flux was monitored only with the aid of the ordinary power reading equipment of the reactor. To reduce the influence of local power fluctuations many successive runs were taken with and without a filter in the beam.

For the fission product activity measurement small plutonium foils (8 mm in diameter, Pu content 2.5 mg/cm^2) were irradiated in the central channel. Due to the large cadmium ratio for Pu239 the bare and the cadmium covered foils were irradiated at different power levels $(P_{Cd}/P_{Bare} \approx 20)$. Lead gold foils were used for monitoring purposes. The cadmium run was made with the foil placed in a small cadmium cylinder (H/D $\approx$ 1) with a wall thickness of 3 mm. A large thickness was chosen to avoid the need for an accurate calculation of the cadmium cut-off energy, E_{Cd}, for plutonium in cylinder geometry and isotropic flux. With a thick filter E_{Cd} falls in a region where the Pu239 fission cross section is low, and large uncertainties in E_{Cd} may be accepted.

The γ-activity from the fission products was measured with a scintillation spectrometer using a single channel pulse height analyzer. The analyzer was either set to correspond to γ energies above 0.2 MeV or to a window covering the energy region 0.2 to 0.3 MeV. The activity measurements started about 18 hours after the stop of the irradiations and points were taken during several days.

The result of this measurement will be briefly summarized in section V.1.

IV.   The neutron spectrum

The accuracy that can be obtained in measurements of the present kind is largely dependent on how well the neutron spectrum is known. For the cadmium filter measurements, yielding the dilute resonance integrals, the thermal and the low energy part of the epithermal region are most important. For the boron filter studies also a good knowledge of the high energy part is required for a meaningful measurement.

IV.1   The unfiltered beam

IV.1.1   The thermal spectrum

The neutron temperature at the centre of the reactor has been determined by two independent measurements. The earlier experiments by Johansson, Lampa and Sjöstrand [7] using a fast chopper gave a neutron energy distribution which well fitted a Maxwellian distribution function with a neutron temperature $T_n = 29\ ^{\circ}C \pm 10\ ^{\circ}C$ above the moderator temperature. Later extensions of the measurements have yielded a somewhat lower and more accurate value, $T_n = 22\ ^{\circ}C \pm 4.5\ ^{\circ}C$ [8]. This value is in good agreement with that obtained in a recent experiment by Sokolowski, Pekarek and Jonsson [9]. By studying the reactor response to spectrum sensitive absorbers such as Cd, Gd and Sm with pile oscillator technique a value of $T_n = 22.5\ ^{\circ}C \pm 3.5\ ^{\circ}C$ could be deduced. For the treatment of our data we assume the excess neutron temperature to be $22\ ^{\circ}C$.

Along the path from the scatterer at the reactor centre to the sample - a distance of about 4.3 m - neutrons are scattered out of (or absorbed in) the beam by collisions against the nitrogen and oxygen atoms in the air. As the cross sections for these substances increase with decreasing energy in the thermal and near thermal region, the spectrum at the collimator exit becomes somewhat distorted. A rough calculation shows that the Maxwellian flux is shifted towards higher energies by an amount corresponding to a change in neutron temperature of about $5\ ^{\circ}C$. The Maxwellian form is not conserved but in the present case it is sufficient to approximate the true flux at the detector position by a Maxwellian with $T_n = 27\ ^{\circ}C \pm 5\ ^{\circ}C$ above the moderator temperature.

IV.1.2   The energy region from the joining region to about 10 keV

For the energy region indicated in the heading, use was made of the fast chopper result by Johansson et al. [7, 10, 11] . The corrections for absorption and scattering in air were made with the data given in [10] . The resulting spectrum is illustrated in Fig. 4.

The curve in Fig. 4 is normalized to 1 at the gold resonance, 4.9 eV. According to Johansson the flux level at any other energy within the region studied is determined relative to the 4.9 eV value to better than $\pm$ 4 %. However, most of the contribution to the resonance absorption comes from energies quite close to the gold resonance. For that energy region the flux level is known to better than $\pm$ 1 %.

IV.1.3   The high energy region

The neutron energy distribution between 10 keV and 1 MeV is not very well known. There exists a Monte Carlo (M.C.) calculated spectrum [12, unpublished] for the centre of the R1 central channel covering the energy region 15 MeV to 25 keV. However, the form of this spectrum below a few hundred keV cannot easily be reconciled with a probable extrapolation of the chopper curve (compare Fig. 5, to be discussed below). Therefore, instead of trying to join the chopper curve to the M.C. spectrum directly a measurement was made of the epicadmium U238 and U235 fission ratio. The fission cross section for U238 is negligibly small below 1 MeV while the main contribution to the U235 fission rate arises from the low energy region. Therefore, the measured ratio could be used for determining the amplitude of the lethargy flux above 1 MeV to that of the chopper curve (compare below).

The experiment was performed in the centre of the central channel rather than in the extracted beam for intensity reasons. Three cadmium covered chambers were lowered into the channel, one with enriched uranium (atom % U235, p_1^{25}, 93.3), the others with depleted uranium (p_2^{25} = 5.9 $\cdot$ 10^{-2} or 2.0 $\cdot$ 10^{-2}). Bias curves were accurately determined for all chambers and the pulse rates were measured at a suitable bias setting. The experiment was then repeated in the thermal column without cadmium around the detectors. By extrapolating the pulse rates to zero bias with the aid of the bias curves, errors caused by electronic drifts, differences in cable lengths for the two runs etc. could be reduced.

The measured pulse rates were related to calculated quantities through the following set of equations.

Central channel:

$$p_1^{25} \, RI' + (100 - p_1^{25}) \, A \int \sigma_f^{28} \, \phi(u)du = k_1 \, N_1^c \quad \ldots \ldots \ldots \quad (1)$$

$$p_2^{25} \, RI' + (100 - p_2^{25}) \, A \int \sigma_f^{28} \, \phi(u)du = k_2 \, N_2^c \quad \ldots \ldots \ldots \quad (2)$$

Thermal column:

$$p_1^{25} \, \phi_{th} \, g \, \sigma_{fo}^{25} = k_1' \, N_1^t \quad \ldots \ldots \ldots \ldots \ldots \quad (3)$$

$$p_2^{25} \, \phi_{th} \, g \, \sigma_{fo}^{25} = k_2' \, N_2^t \quad \ldots \ldots \ldots \ldots \ldots \quad (4)$$

RI'   is the epicadmium fission integral for U235 in the R1 flux

σ_f^{28} is the fission cross section of U238

$g \, \sigma_{fo}^{25}$ is the effective 2200 m/s U235 fission cross section for the thermal column run

$\phi(u)$ is a normalized fission neutron spectrum $(\int \phi(u)du = 1)$ or the M.C. spectrum (ad hoc normalized)

ϕ_{th} is the conventional flux in the thermal column run

A   is the amplitude sought

k_1, k_1', k_2 and k_2' are constants involving efficiencies of the chambers, number of nuclei etc.; $k_1 \, k_2' = k_1' \, k_2$

N_1^c and N_2^c are the pulse rates with the chambers in the central channel

N_1^t and N_2^t are the pulse rates in the thermal column

In Eqs. (1) and (2) the U235 fission rate per nucleus has been put equal to RI', which implies that the lethargy flux at the gold resonance has been normalized to 1 (compare section V.1). Further, in the term expressing the U238 fission rate, A $\phi(u)$ has been substituted for the true flux, A being the amplitude sought and $\phi(u)$ either a fission neutron spectrum or the M.C. spectrum. The integral $\int \sigma_f^{28} \, \phi(u)du$ was obtained by using the group constants given in [13] for the fission spectrum or the cross section data in [14] for the M.C. spectrum.

The index 2 of p, k and N in the expressions above refers either to the one or the other of the two depleted chambers. Two chambers were used merely to get more reliable results. The value of A, obtained from the two detectors, agreed within about 5 %.

With the aid of the measured pulse rates the factor A could be determined from Eqs. 1 - 4 for the fission and the Monte Carlo spectra. The following values were found:

Fission spectrum: A = 1.86 ± 0.14
Monte Carlo   : A = 1.50 ± 0.12

The main contribution to the resulting limits of error originates from the uncertainty in the fission cross section of U238 (± 5 %). The two A-values should not be compared relative to each other as the M.C. spectrum used for evaluating the integral discussed above was quite arbitrarily normalized.

As mentioned earlier, the M.C. spectrum was calculated to a low energy limit of 25 keV. Using the amplitude obtained from the fission ratio experiment the spectrum relates to the chopper curve in the way shown on Fig. 5. At 10 keV - assuming a horizontal continuation of the M.C. curve - there is a discrepancy in amplitude of about 10 - 15 % which is within the common limits of error. However, the chopper curve drops about 6 % from 1 keV to 10 keV. The decrease in $\phi(u)$ with increasing energy will most probably continue above 10 keV. Support for this assumption is given by results from Canadian calculations [16]. Therefore, we disregard the spectral form represented by the M.C. curve in the energy region 10 keV - 1 MeV and assume instead a linear decrease in $\phi(u)$ with ln E, the amplitude coinciding with the chopper curve at 10 keV and at 1 MeV attaining the value 0.85.

The latter value is chosen somewhat arbitrarily. The exact value is not particularly critical for the conventional fission integrals (0.5 eV < E < 1 MeV) as only about 3 % of the total integral originates from energies above 10 keV. For the boron filter results, however, the insufficient knowledge of $\phi(u)$ in this region is more important. Fortunately, the number of fissions per unit lethargy is largest at the low energy end, in which part the uncertainty in the flux is smallest. In estimating the final limits of error we have crudely assumed the contribution to the margin of errors from the energies above 10 keV to be 0.7 - 0.8 b.

Nilsson and Aalto [15] have earlier made some measurements of the high energy flux with different threshold detectors. The M.C. spectrum fitted to the experimental points agrees within a few per cent with that obtained from the U238/U235 fission ratio measurement. Nilsson (private communication) claims the uncertainty in their amplitude of the M.C. curve to be between 10 and 15 %.

The spectrum at the top of the reactor was slightly distorted because of the scattering in air and additional moderation in the graphite scatterer. The former effect was corrected for with the aid of the known cross sections for oxygen and nitrogen. The resulting curve is shown on Fig. 5. For estimating the moderation effect a measurement was made of the U238/U235 ratio close to the centre with and without the scatterer in place. With the scatterer the A value was about 3 % lower than that found without the scatterer. The value should be still lower at the top of the reactor where the detectors were only hit by neutrons emitted from the scatterer. However, compared to the large overall uncertainty in the high energy flux, the effect is small and it has been disregarded in the treatment of the data.

IV.2   The filtered beams

IV.2.1   Cadmium filter

The transmission characteristics of the thin cadmium filter were measured with the fast chopper from thermal energies up to about 50 eV[x]. For the thick cadmium the range was extended to an upper limit of about 2500 eV. Thereby a rough estimate could be made of the effect of cadmium resonances other than the first one, as well as of the reduction of the beam because of scattering in the cadmium.

A transmission curve for the thick filter in the range 0 - 3 eV is given in Fig. 6. The curve is normalized to 1 at about 5 eV.

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[x] All chopper measurements were made by the chopper group headed by E. Johansson.

The effective cadmium cut-off values were obtained from the following relation

$$\int_{0}^{1\ MeV} T(E)\ E\ \phi(E)\ \sigma(E)\ \frac{dE}{E} = \int_{E_{Cd}}^{1\ MeV} \sigma(E)\ \frac{dE}{E} \quad \cdots \cdots \qquad (5)$$

$T(E)$ is the normalized transmission curve for the cadmium sheet used. $T(E)$ is put equal to 1 for energies above 4 - 5 eV. The corrections for absorption in resonances above the first one and scattering are applied directly to the measured cadmium ratios.

$E\ \phi(E) = \phi(u)$ is the unfiltered lethargy flux, normalized to 1 at the 4.9 eV gold resonance.

$\sigma(E)$ is for gold the $1/v$ part of the absorption. For uranium and plutonium it denotes the total fission cross sections. The latter were taken from the Karlsruhe compilation [17].

It is obvious that the E_{Cd} values defined by Eq. 5 depend not only on the cadmium thickness and the energy dependence of the cross sections in question, but also on the properties of the unfiltered beam. The values given below are therefore not directly comparable with those obtained for a true $1/E$ flux.

If $\phi(u)$ is normalized to 1 at a lethargy corresponding to 4.9 eV the deviation from 1 in the actual flux for energies between 2 eV and 1 MeV corresponds to an increase in the fission integral of about 9.0 b for U235 and 10.5 b for Pu239. The contribution to these values from energies above 10 keV is for both materials less than 5 %.

Numerical integration of the integrals given in Eq. 5 yielded the following cut-off energies

Table I

Cadmium cut-off energies

	E_{Cd}		ΔRI to E_{Cd} = 0.5 eV (barns)	
	Cd I[x]	Cd II[x]	Cd I	Cd II
Au	0.497	-	0	
U235	0.446	0.533	- 10	+ 5
Pu239	0.420	0.495	- 53	- 2

[x] Cd I :   thickness 0.105 cm

CdII :   "   0.210 cm

The ΔRI column shows the cross section difference between a 0.5 eV cut-off and the actual one.

The uncertainties in $T(E)$ and $\phi(E)$ yield a resulting error in E_{Cd} corresponding to about 4 b in the resonance integrals, the main contribution arising from the uncertainty in $\phi(E)$.

IV.2.2   Boron filters

In the energy region covered by the chopper ($E <$ about 10 keV) the scattering cross section of the filters is to a good approximation energy independent and the absorption cross section varies as $1/\sqrt{E}$. The transmission of the filters might therefore be written as $T(E) = B \exp(-\beta E^{-1/2})$. As E is inversely proportional to t^2 where t is the time-of-flight ln $T(t)$ should be a linear function of t. In Fig. 7 the measured transmission curve is given. As may be seen a straight line can well be fitted to the measured points. The large spread in the points at low energies is due to poor statistics in this energy region and to uncertainties in the background that has to be subtracted from the measured points.

The constants B and β could in principle both be determined by a least squares fit of $T(E)$ to the experimental values. This was in fact done, but probably due to an uncertainty in the synchronization of the chopper pulses relative to the channels in the time analyzer the B values differed, outside the expected limits of error, from the values calculated from the macroscopic scattering cross sections. Only the β values, therefore, were taken from the least squares calculation and B was determined with the aid of the cross sections.

Table II shows the values for B and β. The errors in β are those given by the least squares calculations. The limits of error attributed to B correspond to an assumed uncertainty in the macroscopic cross section of 10 - 15 %.

Table II

The constants B and β for the ooron filter transmission function

	B	β
Thin filter	0.920 ± 0.015	9.60 ± 0.10
Thick filter	0.850 ± 0.015	23.3 ± 0.2

To calculate the fission rate in the filtered beam the transmission function must also be known for energies above 10 keV. As experimental data of the B^{10} absorption cross section at high energies is quite inaccurate, σ_a has been assumed to follow the $1/v$ law over the whole energy range. This will result in a slight underestimate of the transmitted beam at very high energies, as available data [18] indicate a more rapid decrease of σ_a with energy than $1/v$. In addition, the variation of the scattering cross section with energy should be considered. σ_s for B^{10} decreases somewhat with increasing energy. The effect of this is, however, compensated for by the increase in the effective cross section of the aluminium in the walls of the ooxes containing the boron (aluminium thickness for the thin filter was 3 mm, for the thick one 2 mm). The factor B will exhibit rapid variations in the energy regions where aluminium resonances appear. The error in considering B and β constant over the whole energy region is, however, small compared to the uncertainty in $\phi(u)$. The same B and β values have therefore been used over the entire energy range.

V.   Results

V.1   Dilute integrals

The cadmium ratios obtained from the different runs are summarized in Table III. The uncertainties in the correction for scattering and absorption above a few eV in the cadmium filter are included in the errors quoted.

Table III

Measured cadmium ratios for gold, U235 and Pu239

Substance	R_{Cd}	
	Cd I	Cd II
Au	2.262 $\pm$ 0.018	-
U235	38.9 $\pm$ 0.3	41.6 $\pm$ 0.5
Pu239	48.7 $\pm$ 0.4	58.6 $\pm$ 0.5

The cadmium ratio, $R_{Cd}(x)$, for a substance, x, may be written

$$R_{Cd}(x) = \frac{\hat{\sigma}(x)}{r\sqrt{\frac{4T}{\pi T_o}}\, RI(x)} \quad \ldots \ldots \ldots \ldots \quad (6)$$

$\hat{\sigma}(x) = \sigma_o(x)\,[\,g(T) + r\,s(T)]$ is the effective cross section for x in a neutron spectrum characterized by the neutron temperature T ^oK and the epithermal fraction r.

$RI(x)$ is the resonance integral for x including the $1/v$ part.

Eliminating the factor $r\sqrt{\frac{4T}{\pi T_o}}$ with the aid of the cadmium ratio for gold the following expression is obtained for the unknown resonance integral

$$RI(x) = \frac{\hat{\sigma}(x)}{\hat{\sigma}(Au)} \cdot \frac{R_{Cd}(Au) \cdot RI(Au)}{R_{Cd}(x)} \quad \ldots \ldots \ldots \ldots \quad (7)$$

The cross sections used were

RI(Au)	= 1550 $\pm$ 35 b (based on a dilute integral of 1510 $\pm$ 35 b [6, 19, 20], a self shielding correction of 5 b and a $1/v$ part of 45 b)
σ_o(Au)	= 98.4 $\pm$ 0.5 b [21]
σ_o(U235)	= 582.2 $\pm$ 2.2 b [22]
σ_o(Pu239)	= 748.2 $\pm$ 4.9 b [22]
r	= 0.0411 $\pm$ 0.0011 as determined from the gold measurement and the assumed neutron temperature.

The g and s (s_4) values were taken from Westcott's compilation [1]. The epicadmium fission integrals on which a part of the s_4 values are based are close enough to those obtained in our measurement not to require any recalculations.

Using Eq. 7, the values in Table III and the data given above, RI(x) for U235 and Pu239 were determined. Before being quoted as fission integrals the effective contributions from energies above 1 MeV were subtracted. The contributions in question - about 1.9 b for U235 and 2.9 b for Pu239 - were calculated using the Monte Carlo spectrum discussed earlier and the cross section data in [17] . In addition, a correction was made to refer the integrals to a standard cut-off of 0.5 eV. The following values were then obtained:

$$U235 \quad : \quad RI = 278 \pm 9 \text{ b}$$
$$Pu239 \quad : \quad RI = 301 \pm 10 \text{ b}$$

The values are weighted means from the runs with different cadmium thicknesses.

The measurements on the plutonium foils yielded a cadmium ratio of 68.3 ± 1.5. It turned out to be independent of the γ energy region chosen for the measurement as well as of the time, t, after the stop of irradiation (measurements were made at t = 18 hours, 3 and 12 days for the run with E_γ between 0.2 and 0.3 MeV, and at t = 3, 4 and 12 days with $E_\gamma > 0.2$ MeV). The experiment only served the purpose of crudely comparing the resonance integrals from fission chamber and fission product activity measurements. Therefore, it was not considered worth while to make an accurate calculation to obtain the cadmium cut-off energy for this case, in which isotropic flux was used. However, E_{Cd} should lie between the value valid for a 1/v absorber inside the cadmium box and placed in an isotropic flux and that obtained for Pu239 behind a 3 mm sheet of cadmium in beam geometry. The former E_{Cd} value, obtained by extrapolation from tables in [23] , yields a resonance integral of 302 b, while the use of the latter (obtained by numerical integration) gives 287 b. For the present purpose the mean value between the two extremes is an acceptable choice. Allowing, crudely, 3 b (1 %) for shielding effects because of the cadmium resonances above the first one a value of 298 ± 9 b is obtained. The uncertainty quoted is only that relative to the result from the fission chamber measurement. Thus, the resonance integrals of Pu239 obtained with the two different methods of measurement are in close agreement.

The main contribution to the limits of error quoted for the fission chamber results originates from the uncertainty in the resonance

integral of gold. Other errors that have been considered are uncertainties in the lethargy flux, in the cadmium cut-off, in the cadmium ratios involved, and in the other cross sections used.

V.2   Comparison with results from calculations and other measurements

Table IV contains the results from calculations and earlier measurements. Besides the original integrals there is also a column with the values corrected to a common cut-off energy of 0.5 eV. All results include the $1/v$ part.

Table IV

Comparisons between resonance integral values
obtained in different experiments

U235

Reference	E_{Cd}	RI	RI $E_{Cd} = 0.5$ eV	Method
Hardy et al. [3]	0.50	274 ± 11	274 ± 11	F.P.[x]
Baumann [4]	0.60	263 ± 9	276 ± 9	F.P.
Clayton [2]	0:49	271 ± 25	269 ± 25	F.C.[x]
Bigham [5]	0.45	272 ± 8	263 ± 8	F.C.
Present work	0.50	278 ± 9	278 ± 9	F.C.
Calculated			269^{xx}	

Pu239

Reference	E_{Cd}	RI	RI $E_{Cd} = 0.5$ eV	Method
Hardy et al. [3]	0.50	327 ± 22	327 ± 22	F.P.
Bigham [5]	0.45	385 ± 18	360 ± 18	F.P.
Bigham [5]	0.45	314 ± 9	289 ± 9	F.C.
Present work	0.50	301 ± 10	301 ± 10	F.C.
Calculated			298^{xx}	

[x] F.C. = fission chamber measurement

F.P. = fission product activity measurement

[xx] Based on data given in KFK-120, I-III, compare text.

The integrals obtained in the present work are larger than those obtained by Bigham in his experiments with fission chambers. As parts of the limits of error are common to both measurements, it appears as if the difference were significant. It is interesting to note that the ratio of the quoted fission integrals for U235 and Pu239 for the two experiments is the same within about 1 %. Bigham used lithium, boron and gold as standards, thereby reducing somewhat the uncertainty caused by the cross section standard. On the other hand the spectral conditions were less favourable in his experiment as the neutron beam originated from the outer surface of an inner reflector. The neutron energy distribution therefore deviated quite considerably from 1/E, requiring the application of large correction factors.

The calculated values given in Table IV are based on cross sections listed in the Karlsruhe compilation, KFK-120 [17] . For U235 the data were taken directly from part II of the reference quoted. The result obtained, 269 b, is probably accurate to about 5 %. All experimental and calculated values for U235 fall within the common limits of error.

For plutonium the Karlsruhe compilation, parts II and III, was originally used. As pointed out by J.J. Schmidt [24] , however, the cross section values around the 14.68 eV resonance given in these volumes are in error. New data will be given in KFK-120, Part I. Using the corrected data the resonance integral, 298 b, given in Table IV is obtained.

For U235 no systematic differences between results from fission product (F.P.) and fission chamber (F.C.) measurements are observed. Thus, within the neutron energy region of importance for the resonance absorption there seems to be very small variations in the yields of the fission products of interest for the F.P. measurements.

Similar agreement is found for Pu239 between the F.P. value reported by Hardy et al. [3] and our F.P. and F.C. results. Bigham's two values, on the other hand, differ more than 20 %. However, his F.P. and F.C. measurements were not performed in the same spectrum (the foil irradiations were made at a cell boundary in a ZEEP lattice). In view of the close agreement found for our F.P. and F.C. results it might be, that part of the difference, found by Bigham, is due to incorrect assumptions concerning the neutron spectra used in his experiments.

V.3 "Boron filter" integrals

As indicated in the introduction the objective of the boron filter runs was to get a check of the fission cross sections above the immediate proximity of the cadmium cut-off energy. Because of the particular characteristics of the fission resonances, especially the close average spacing, the contributions to the measured pulse rates in the filtered beams are not concentrated to a few discrete energies but are fairly evenly distributed over a large energy region. This is well illustrated on the histograms given on Figs. 8 and 9 calculated from the cross section data in [17] and the measured transmission functions. For the thin filter about 50 % of the fissions occurs below 250 eV for U235 and below 125 eV for Pu239. For the thick filter the corresponding figures are 900 eV for U235 and 750 eV for Pu239. With thicker filters the median energy would move further up in energy but the uncertainty in the lethargy flux at the high energy end of the spectrum would make the measurements less meaningful.

It is obvious that for the materials studied the boron filter method cannot give any detailed cross section information. However, for some other substances it may be of more importance. It is particularly interesting to note that in taking the difference between results from a thin and a thick filter run the high energy contribution disappears. Using the boron filter technique on a material with large level spacing it should, in principle, be possible to study the absorption in a single resonance or a group of resonances apart from the absorption at higher energies. In sodium, for instance, the first resonance is at 2.85 keV, the next one at 54 keV. With suitably chosen filters the difference would contain considerable contributions from the $1/v$ part and the first resonance but only a small fraction of the absorption at higher energies.

The ratios of the fission pulse rates in the unfiltered beam to that in the boron filtered ones are given in Table V.

Table V

"Boron filter" ratios for U235 and Pu239

	Thin B^{10} filter	Thick B^{10} filter
U235	199.5 ± 1.5	476 ± 4
Pu239	331 ± 2	862 ± 6

Effective integrals, RI_{eff}, can be deduced from an expression equivalent to Eq. 5. The results obtained are given in Table VI together with calculated values. The latter ones were derived from the expression

$$RI_{eff} = B \int_o^{1 \text{ MeV}} e^{-\beta/\sqrt{E}} \; E \; \phi(E) \; \sigma_f(E) \; \frac{dE}{E} \quad \cdot \cdot \cdot \cdot \cdot \cdot \cdot \cdot \quad (8)$$

The B and β values are those listed in Table II.

$E \; \phi(E)$ is the lethargy flux normalized to 1 at the gold resonance.

$\sigma_f(E)$ is the fission cross section for U235 and Pu239 respectively.

Table VI

Experimental and calculated values of the effective fission integrals in the boron filtered beams

	Thin B^{10} filter		Thick B^{10} filter	
	Exp.	Calc.	Exp.	Calc.
U235	55.1 ± 2.4	56.0	22.2 ± 1.4	24.8
Pu239	51.1 ± 2.2	50.8	18.2 ± 1.2	20.2

The contribution to the limits of error originating from the insufficient knowledge of the neutron spectrum is estimated to be about 1.6 b for the thin filter and about 1 b for the thick one. Other main sources of error are the uncertainty in the cross section standard and in the constants B and β of the boron filter transmission functions.

By analogy with the resonance integral calculations the upper energy limit in the integral in Eq. 5 has been put equal to 1 MeV. The contribution to the experimental values from energies above 1 MeV has therefore been subtracted before the results were listed in Table VI. The contributions are the same as those given in section V.1 except for the correction for the scattering in the filters.

As is seen from Table VI the results from experiments and calculations are in good agreement for the thin filter. For the thick one the calculated values are about 10 % larger than the experimental ones. However, as an uncertainty of 5 %, at least, should be attributed to the fission cross sections used for the calculations the common limits of error will cover the discrepancy quoted.

VI.   Acknowledgements

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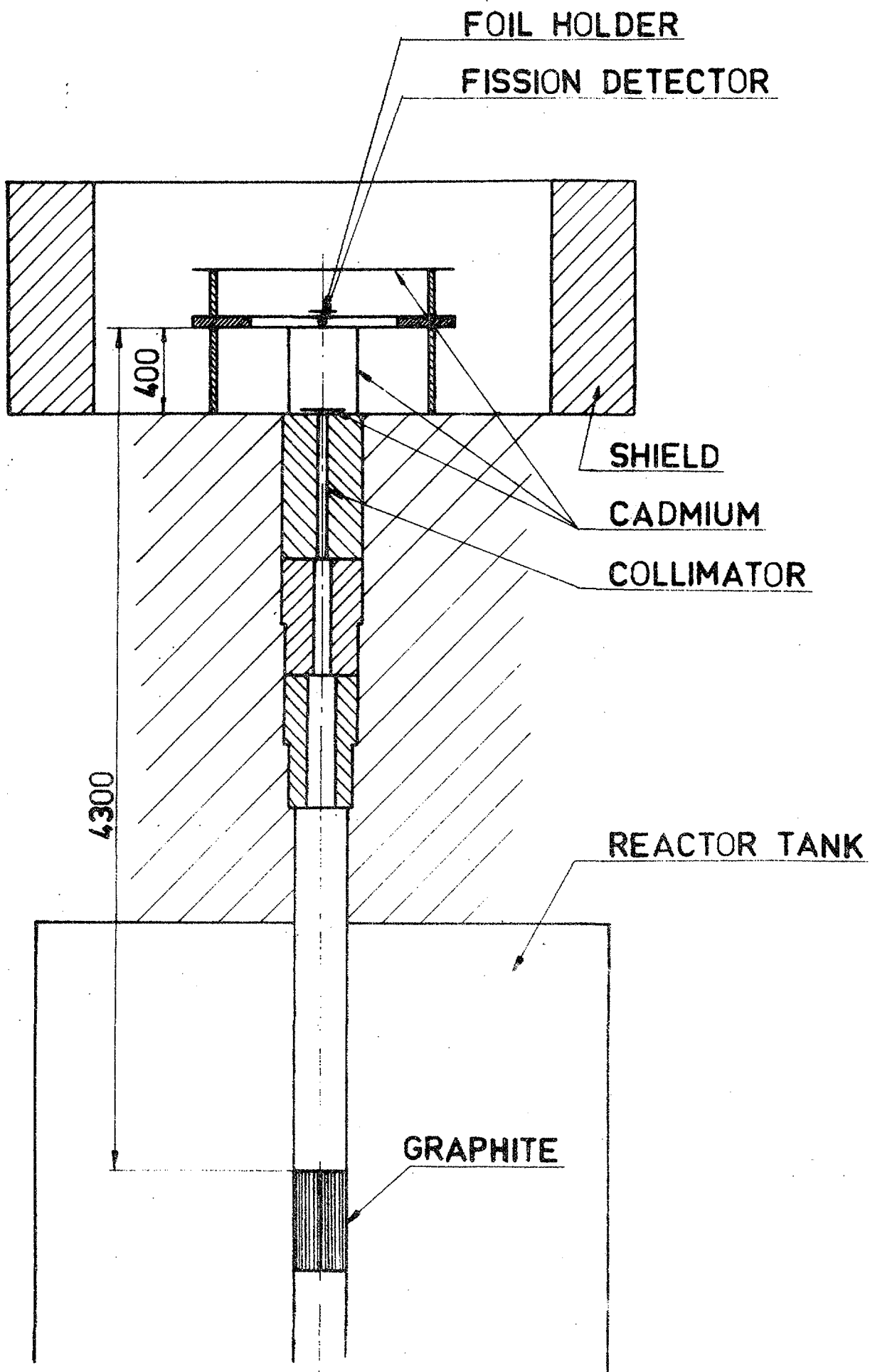COATED LAYER
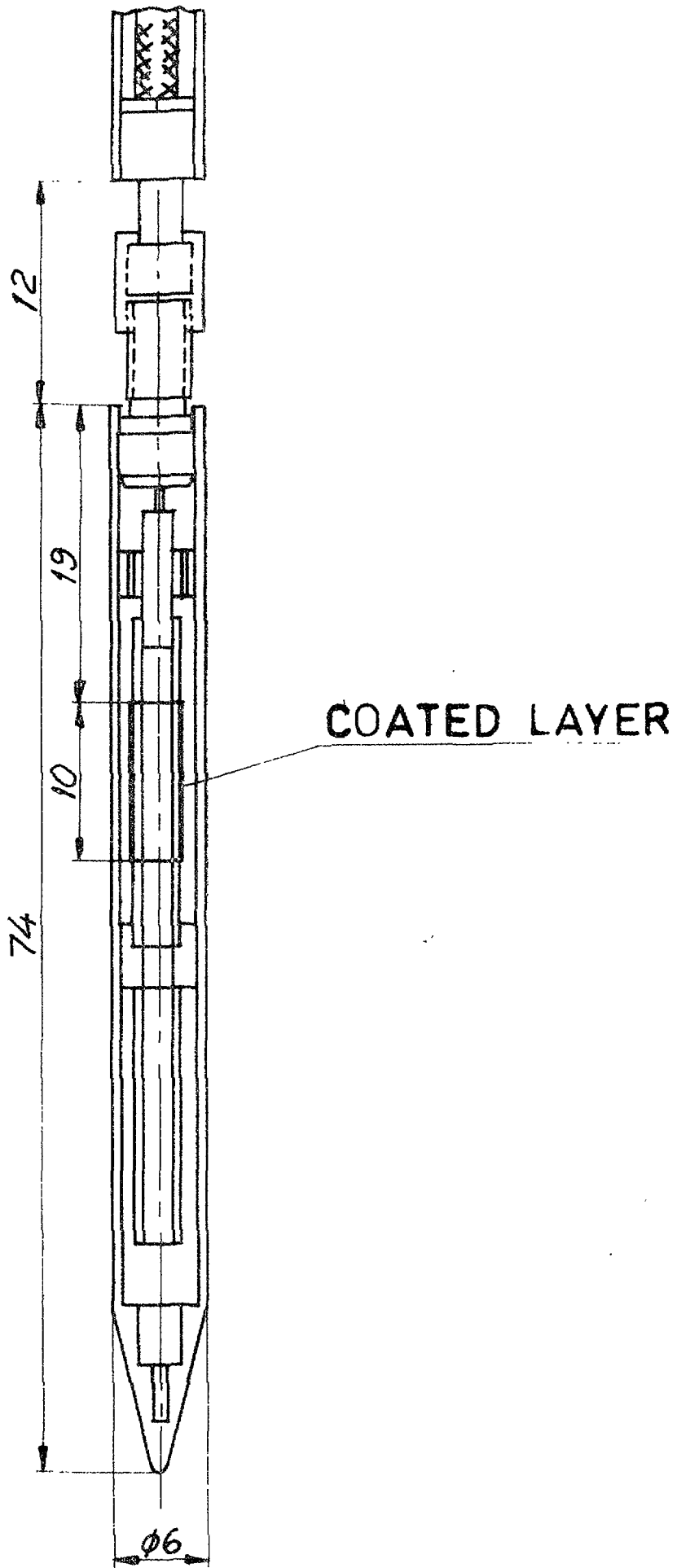
Fig. 1

Dimensions of the fission chambers
(all units in mm)

Fig. 2

Experimental arrangement at the R1 reactor
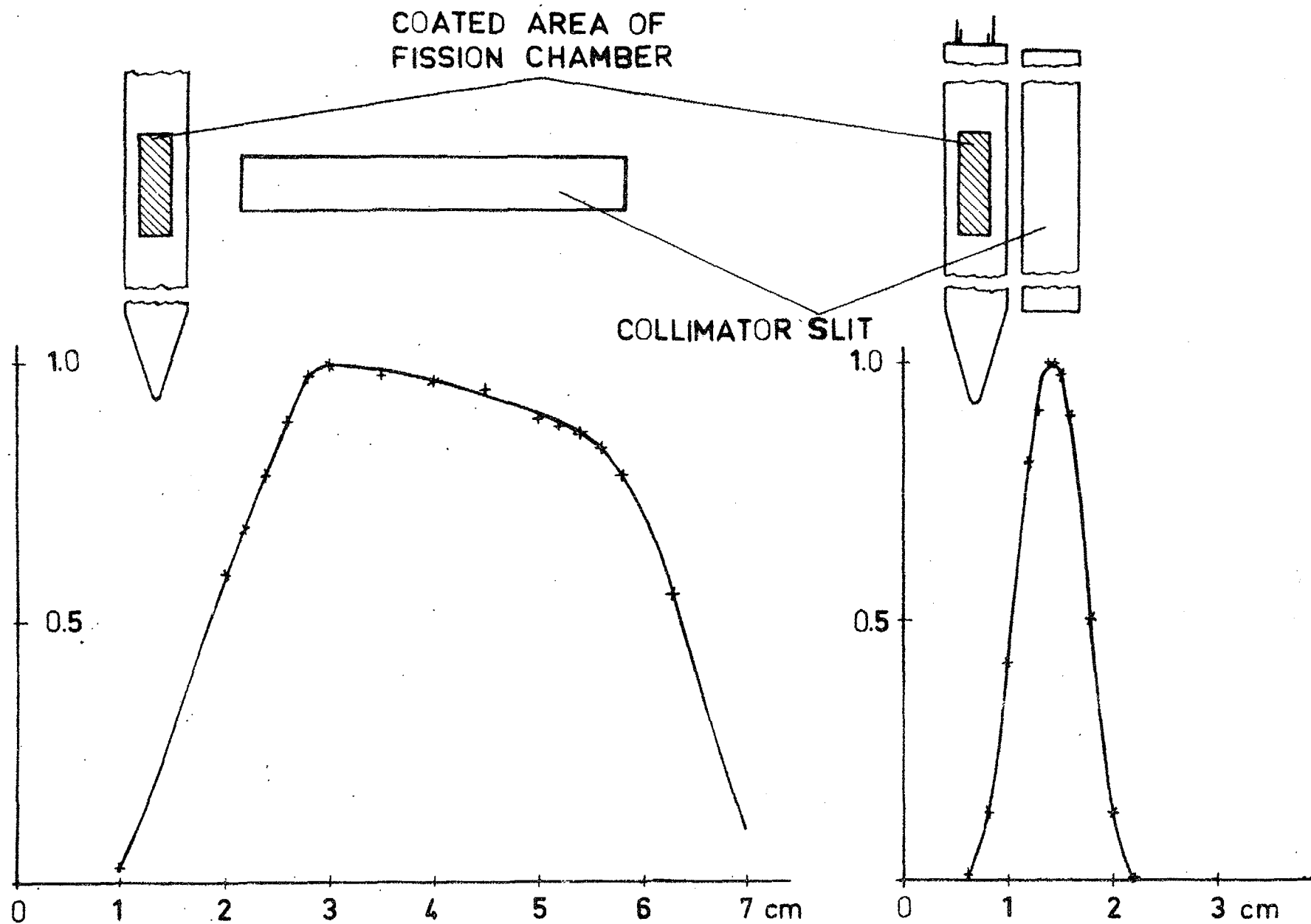
COATED AREA OF
FISSION CHAMBER

COLLIMATOR SLIT

Fig. 3

Beam profile at the position of the fission chambers. The
asymmetry of the curve to the left is most probably due to
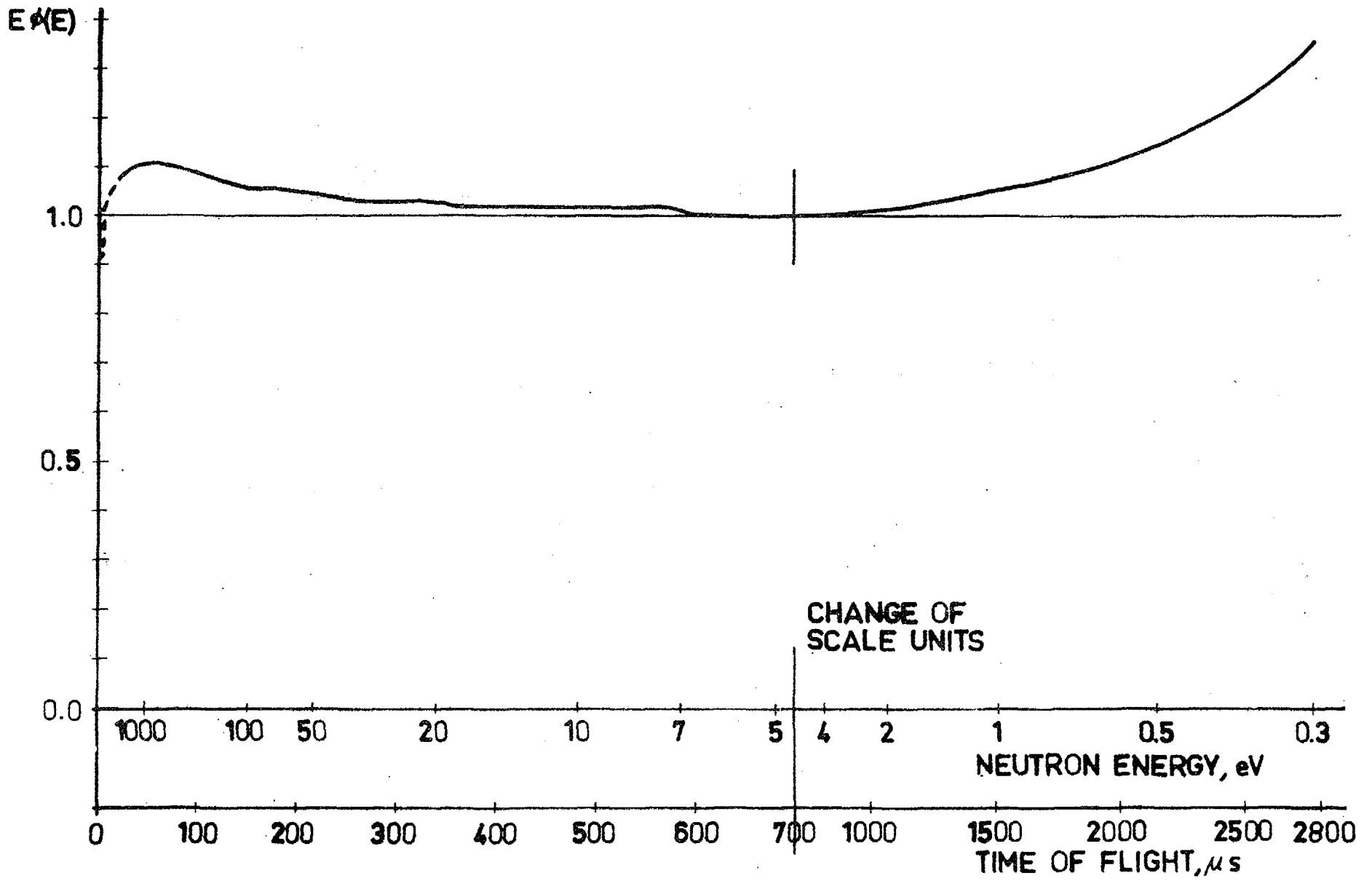an incorrect positioning of the detector.

Fig. 4

The lethargy flux $\phi(u) = E \phi(E)$ in the energy interval 0.3 eV - 10 keV
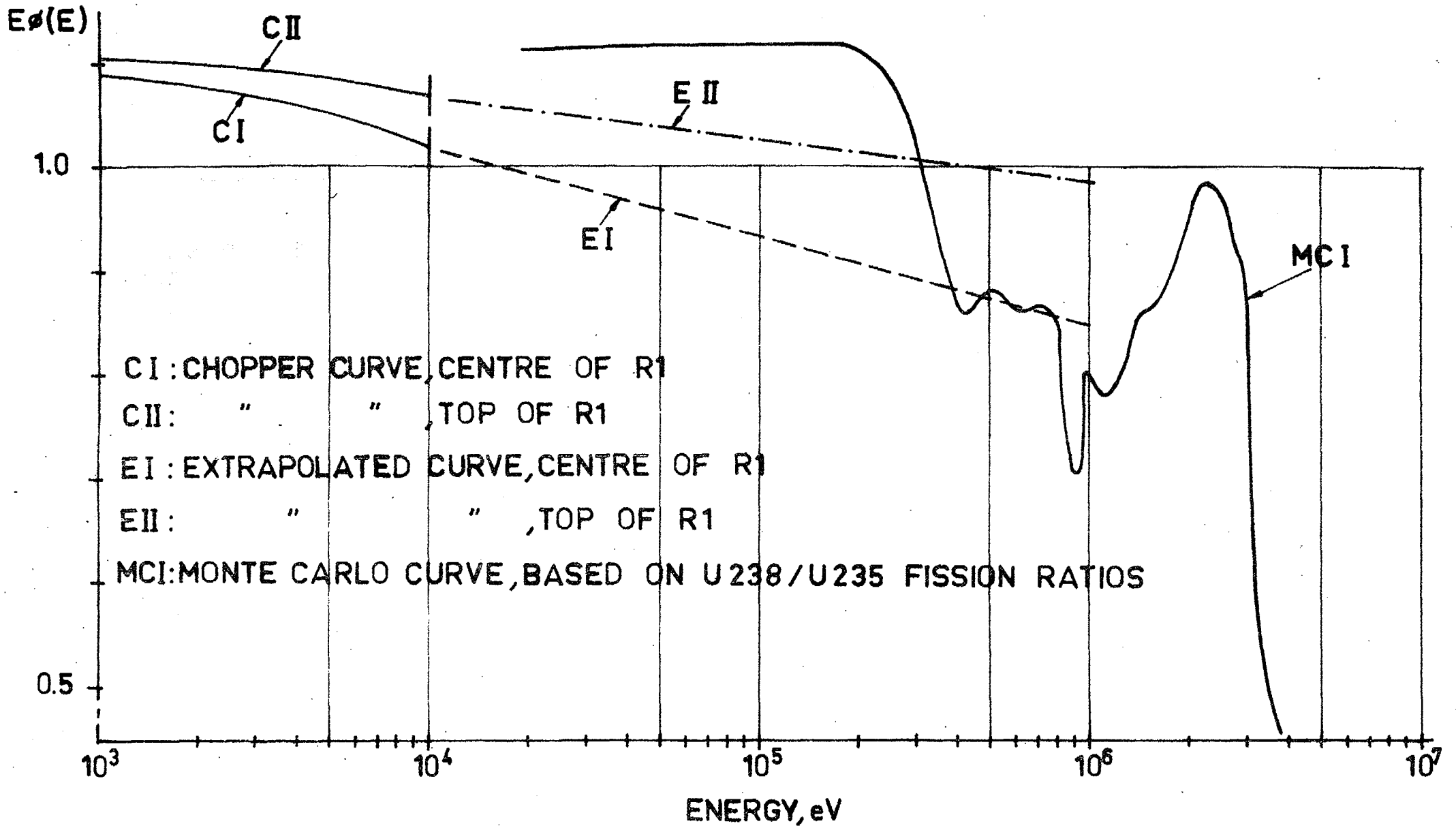
Fig. 5

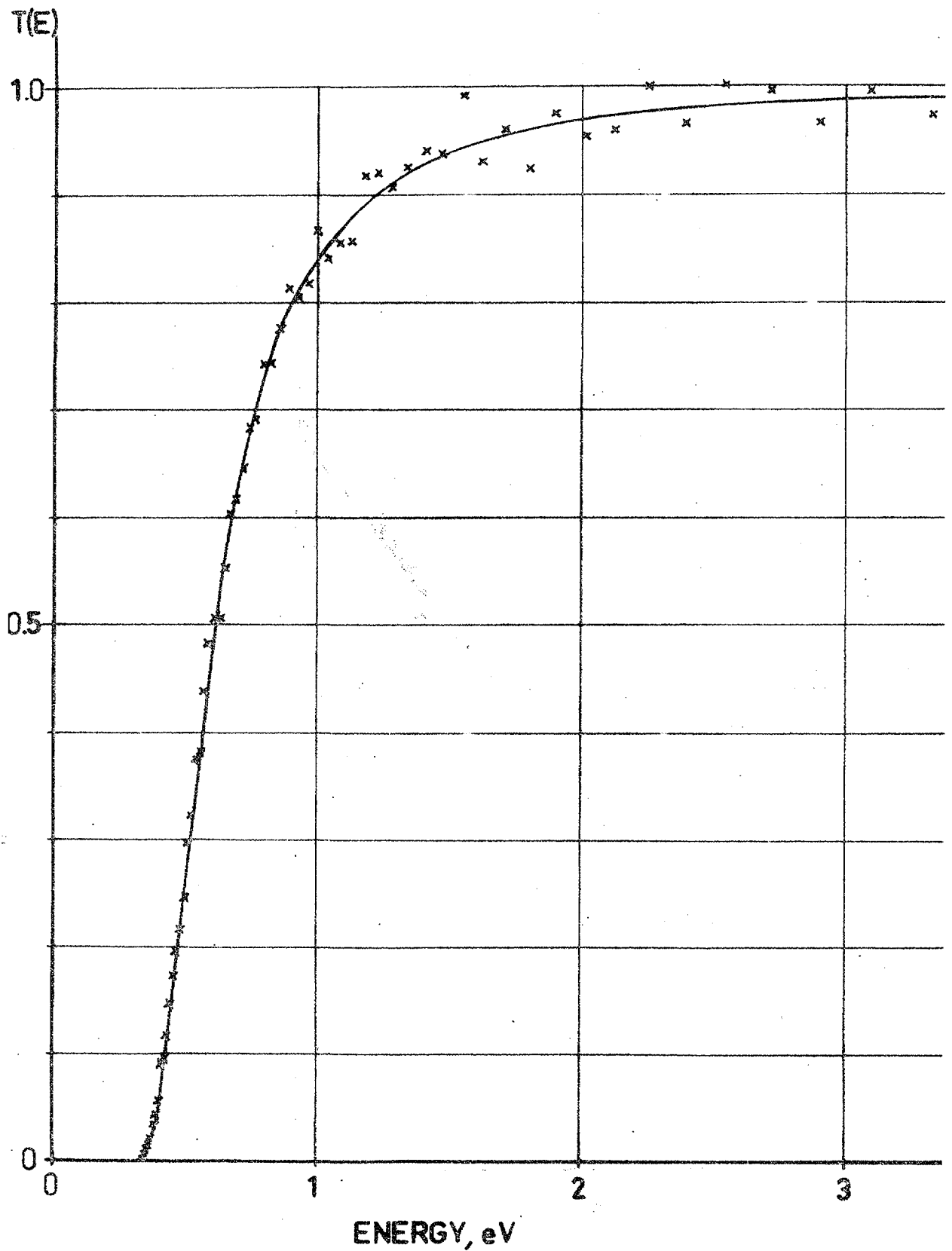The lethargy flux for energies above 1 keV

Fig. 6

The transmission T(E) for the thick cadmium filter.
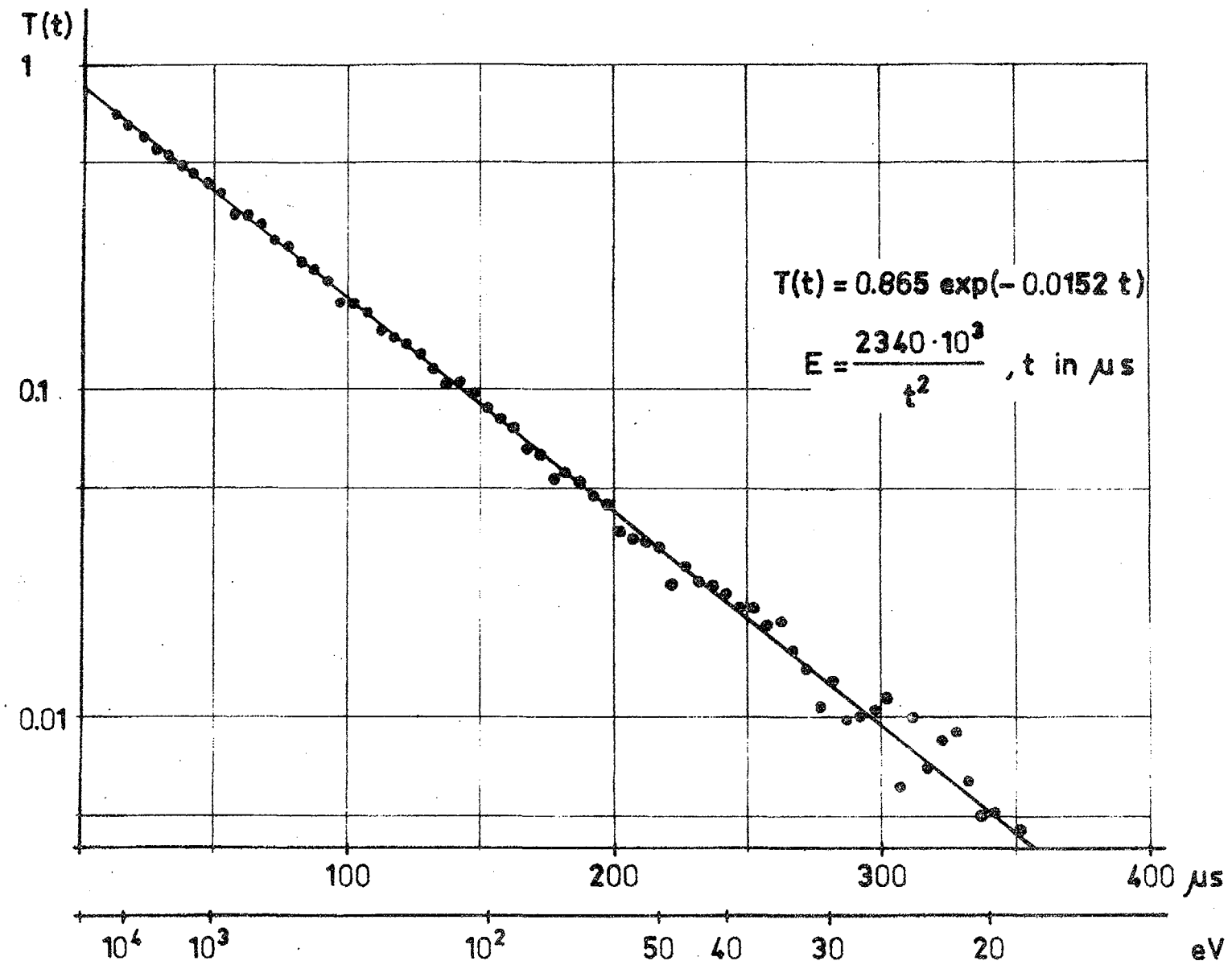T(E) is normalized to 1 at 4 - 5 eV

Fig. 7

The transmission function $T(t)$ for the thick boron filter as a function of time-of-flight. The relation between the neutron energy and time-of-flight in the present experiment was $E = 2340 \cdot 10^3 \cdot t^{-2}$

$$\triangle RI = \int_{E_i}^{E_{i+1}} T(E) E \phi(E) \sigma(E) \frac{dE}{E}$$

$$\ln E_{i+1} - \ln E_i = 1$$

— — — — $T(E) = 1$

———— $T(E) = \exp(-9.59 E^{-1/2})$
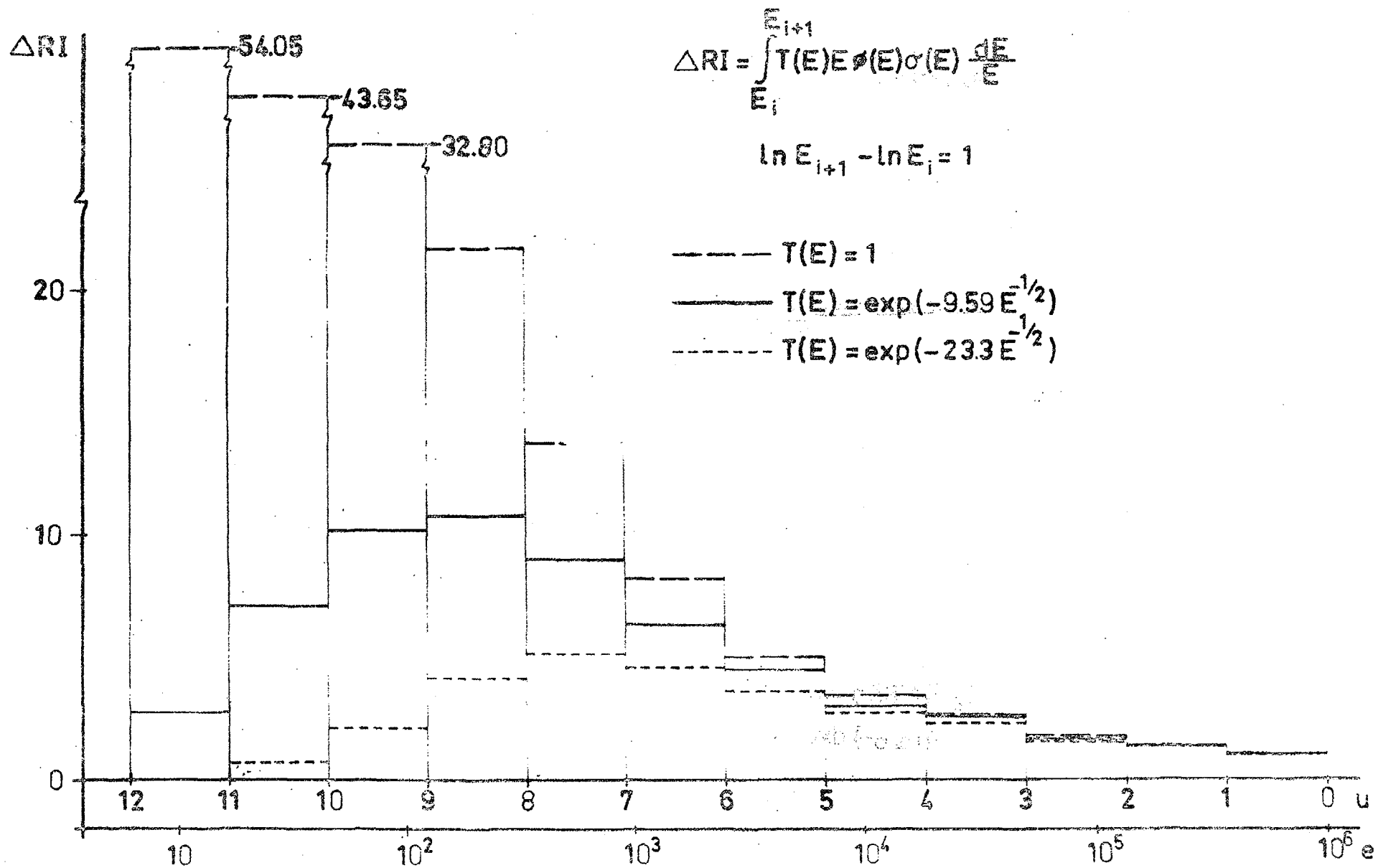
- - - - - - $T(E) = \exp(-23.3 E^{-1/2})$

Fig. 8

The contributions to the U235 resonance integral from different lethargy intervals in beams filtered with cadmium and boron
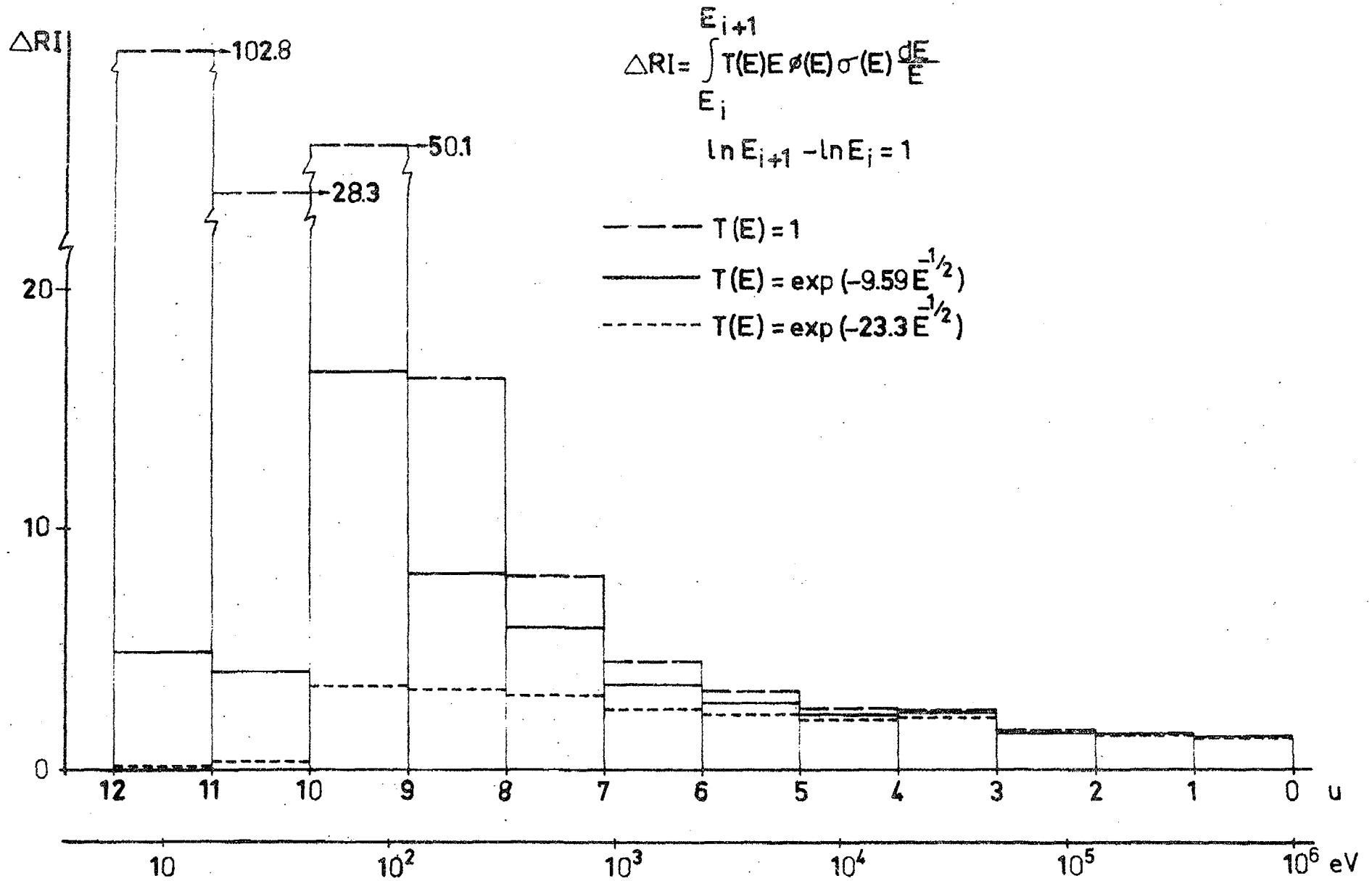
Fig. 9

The contributions to the Pu239 resonance integral from
different lethargy intervals in beams filtered with
cadmium and boron

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