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# SHAPE OF THE $^{238}U$ NEUTRON CAPTURE CROSS SECTION IN THE RANGE 0.002-0.1 eV

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Abstract: The relative neutron capture cross section of $^{238}U$ has been measured in the range 0.002-0.1 eV using a short flight path of the Geel linac and a special liquid methane moderator. The results are in keeping with an approximate 1/v dependence of this cross section and disagree with the surmise of Santamarina et al., who foresee a resonance at $E_o$ = -0.005 eV.

($^{238}U$, capture cross section, range 0.002-0.1 eV)

## Introduction

It has been observed since several years that the interpretation of integral experiments performed on thermal reactor cores give rise to values of temperature coefficients of reactivity which are systematically higher than those calculated from the known microscopic cross sections. Since this discrepancy has been noticed in a variety of systems, from experimental zero-power reactors to those employed in nuclear power stations, and using different calculational methods, it was natural to ascribe it to a deficiency in the basic nuclear data. Several evaluators[1-6] have therefore proposed variations of the shape of certain important cross sections at thermal energy in order to account for the observed effects. The most complete work in this field has been recently carried out by Erradi and Santamarina[4-6] who have been able to reproduce the results of a large variety of integral experiments by introducing negative energy resonances with appropriate parameters in the evaluation of the nuclides $^{235}U$ and $^{238}U$. In particular, for $^{238}U$ a resonance at $E_o$=- 0.005 eV was assumed, contributing 0.2 b to the 2200 m/s cross section. This level makes the capture cross section decrease with energy more rapidly than 1/v, which is the shape currently accepted in all previous evaluations. The aim of the present work is to test experimentally the hypothesis of refs. 4-6 by measuring the shape of the $^{238}U$ capture cross section at thermal and subthermal energy.

## Experimental Procedure

It is clear that the resonance proposed by Santamarina, with $E_o$=-0.005 eV and $\Gamma \cong 23$ meV (according to the average capture width in $^{238}U$) will mainly influence the energy region below 20-30 meV. In order to look for its presence, it is therefore mandatory to be able to measure with reasonable precision down to a few meV energy, a thing which can only be achieved by using a cold moderator. For this reason a cryostat filled with liquid methane at a temperature of about 110 K was placed around the rotary U target of the Geel linac (see Fig. 1). It consisted of two inter-connected boxes of approximate size 13x8.5x7.5 cm having two vacuum isolated stainless steel walls of thickness 0.15 cm each. This container was connected through super-isolated pipes to a liquid methane "factory" situated outside the target room. The methane vapour produced on top of the moderator as a result of γ-heating, estimated to contribute 60 W power, was first compressed and then circulated through a liquid nitrogen heat exchanger where it was liquified again and sent back to the moderator. Operation of this cold moderator resulted in an increase of about a factor 5 in the production of neutrons below 20 meV as compared to the room temperature water moderator normally employed[8]. The layout of the experiment is shown in Fig. 1. The Geel linac was operated to provide bursts of 100 MeV electrons with a current of 0.2 A and a width of 2.2 μs at a repetition frequency of 40 Hertz. The neutron beam and associated γ-ray field were gradually collimated to a 5 cm diameter by placing several collimators of borated wax and lead inside the bunker wall and immediately afterwards. The sample, surrounded by a 0.6 cm thick $^{6}Li$ sleeve, was contained in an evacuated aluminium pipe: it consisted of a thin plate of highly depleted (99.999%) uranium metal of size 6x6 cm and weight 13.8 g, whose thickness was equivalent to $N=0.968 \cdot 10^{-3}$ at/b. It was viewed by four $C_6D_6$ liquid scintillators of 10.2 cm diameter and 7.6 cm height, housed in a large shielding facility with walls made of 14 cm thick lead and 25 cm thick borated wax. The time dependent background was measured by replacing the sample with an equivalent graphite scatterer. The neutron flux at the position of the uranium sample was measured in a separate run by replacing the $^{238}U$ with a sintered $B_4C$ disk of 0.06 cm thickness and 8 cm diameter enriched to 93% $^{10}B$. The 478 keV γ-rays from the reaction $^{10}B$ (n,αγ) were detected with the same $C_6D_6$ scintillators. The constancy of the flux during the various runs was checked by a double gridded ionization chamber placed in the beam before the shielding and loaded with back-to-back deposits of $^{10}B$ of 5 $\mu g/cm^2$ thickness. The signal-to-background ratio of the capture run was improved by a factor 5 by counting only coincidences between γ-rays depositing more than 0.15 MeV in any two detectors. In this way it was possible to achieve a ratio of 16:1 between the peak counting rate per time-of-flight unit, obtained at about 25 meV, and that at 0.8 meV neutron energy where the flat background was mainly due to sample activity. In a second measurement campaign the metallic sample was replaced by a 59.8 g sample of $U_3O_8$, of thickness $N=2.56 \cdot 10^{-3}$ at $^{238}U$/b, also depleted to 99.999% and canned in a thin wall aluminium container of 8 cm diameter.

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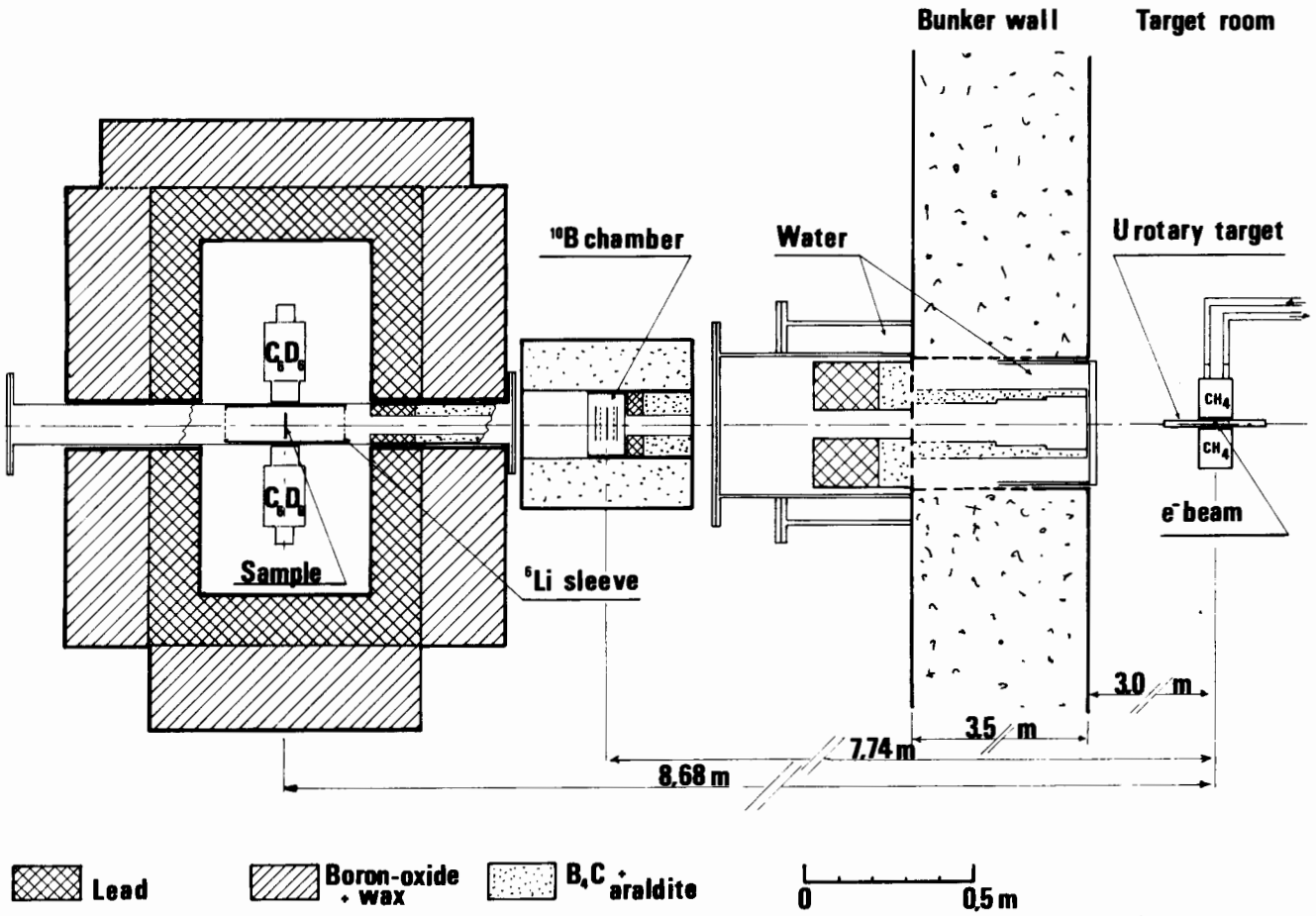


Fig. 1 Vertical cross section of the experimental set up. The electron beam direction is perpendicular to the plane. The thickness of the bunker wall and its distance from target are not in scale

This material was measured immediately after having been chemically purified[9] so that the amount of radioactive decay products, in particular, $^{234}$Th was strongly reduced with respect to the secular equilibrium case; this reduction in activity was however partly compensated by an increase of the energy dependent background mainly due to neutron capture in the container. As a consequence, the peak-to-background ratio defined previously reached a value of 22:1. Both samples were chemically pure to about 99.9%: amongst the several impurities detected by spark source mass spectrometry none contributed an appreciable fraction of the thermal neutron capture in the sample. Results will be given for both samples.

## Analysis

The various steps of the analysis and the related systematic uncertainties are separately discussed in the following:

a) coincidence counting.

Besides having fixed spin and parity ($J^\pi = 1/2^+$), the compound $^{239}$U states contributing to thermal neutron capture decay to the ground state through a large number of open channels in such a way that the influence of Porter-Thomas fluctuations present in single primary γ-rays will be largely smoothed out in the resulting total capture spectrum.Since, as a consequence, the shape and in particular the multiplicity of this γ-spectrum will change very little from resonance to resonance, the coincidence rate of capture γ-rays will represent well the actual capture rate.
In order to experimentally check this point, the coincidence rate C for twenty-two $^{238}$U s-wave resonances in the range 6-478 eV was compared to the corresponding one S obtained in a singles measurement to which the pulse height weighting technique was applied. The same detector setup and metal sample were used but obviously with different Linac parameters and with the normal water moderator. It was found that the relative standard deviation of the C/S population was 2.7% and that all values were included in the interval spanning ± 6% from the average. In the present thermal capture case, the only variation with neutron energy of the γ-spectrum can be due to the $E_o$=-0.005 eV resonance whose relative contribution varies from about 1% at 0.100 eV to 41% at 0.002 eV. Therefore the maximum systematic error associated with the coincidence measurement is ε=0.4x6%=2.4%. This error has been considered over the whole energy range.

b) determination of the background

Two types of background are considered and separately subtracted: the first is a time dependent background which is measured by replacing the $^{238}$U sample with an equivalent graphite scatterer plus, in the case of the U-oxide, a dummy aluminium can. The relative contribution of this background is minimum at the energy of 0.025 eV (typically 5% for the metal and 10% for the oxide sample) and increases with

decreasing energy up to 20-25% at 0.002 eV. The higher value for the oxide sample is due to capture in the canning. A few runs of this type were collected and compared: taking into account fluctuations in their results and normalization errors, we estimated a systematic uncertainty of ± 10% for this kind of background. After subtracting such a contribution, the only background left should be the one associated with the $^{238}U$ sample, which we assume constant with time: this was measured during the main capture run at time-of-flights corresponding to the energy interval 0.65-0.78 meV, where the neutron flux is negligible. The systematic error associated with this type of background depends on the degree of validity of the main assumption, namely that it is constant with time at least in the energy interval of interest. Unfortunately it is impossible to test this assumption, but we notice that the measured background is about 20% larger than that due to sample activity alone, probably because of the influence of sample activation. If we estimate, rather conservatively, that this extra counting rate does not change more than ± 50% below 100 meV, we derive an uncertainty of ± 10% also for this second type of bakground. The fraction of total counting rate due to background varies greatly with energy: in the case of the oxide sample it is 14% at 25 meV, 17% at 10 meV and 73% at 2 meV. It follows that background subtraction contributes by far the largest uncertainty at low energy.

c) Neutron flux

The boron carbide disk used for flux determination is totally absorbing over the whole energy range of interest since its thickness corresponds to $N \simeq 6.2 \times 10^{-3}$ atoms $^{10}B/b$. The detection efficiency is therefore considered independent of energy. On the other hand, the thin layer $^{10}B$ ionization chamber placed in front of the big shielding is only used for monitoring the neutron flux shape and for correcting for possible variations, if needed. In fact, this was not necessary because the flux stayed remarkably constant during the whole experiment. No systematic error was associated with flux determination.

d) Multiple scattering

A correction for multiple scattering was calculated for the oxide sample using a Monte Carlo code. It was found that 4.5% of the captured neutrons have undergone at least one scattering before capture. As expected, this percentage does not change with energy, so that this effect does not play any role as far as the shape of the capture cross section is concerned.

## Results and discussion

The measured values of $\sigma_\gamma \sqrt{E}$ for $^{238}U$ are given in Figs. 2a and 2b for the metal and for the oxide sample, respectively, and compared with the predictions of the available evaluations. Error bars are obtained by combining quadratically the statistical errors and the systematic uncertainties discussed in the previous section. Since the goal here is to compare only the shapes of experimental and calculated curves and not the absolute values, it is found convenient to makeall curves coincide at the upper energy end so that one can look how much the data diverge (or converge) on the low energy side. In absolute terms this is not exact because the two evaluations are normalized to the same value $\sigma_\gamma$=2.70 b of the 2200 m/s cross section. To take that into account, the JEF-1 curve [10] is referred to the left hand vertical scale and the Santamarina evaluation to the right hand one. The JEF-1 curve (full) is in fact representative of any evaluation which does not foresee a negative energy resonance in the immediate vicinity of zero. It has practically a 1/v behaviour except for a 1% slope in the considered energy range due to the influence of the 6.67 eV resonance. The surmise of Santamarina et al. [4-6] on the contrary exhibits a very different slope. An inspection of Figs. 2a and 2b shows that, for both samples, the experimental points disagree with this last evaluation but agree within the errors with the JEF-1 curve. Therefore the assumption of a resonance at $E_o$=-0.005 eV should be dropped. It is interesting at this stage to calculate the probability that such a level exists. In order to contribute 0.2 b to the thermal cross section, this resonance must have $\Gamma_n^\circ = 2.16 \times 10^{-9}$ eV. Taking the $^{238}U$ average resonance parameters from ref.11, the probability for an s-wave resonance of having $\Gamma_n^\circ$ smaller or equal to that value is $P_1 = 0.74 \times 10^{-3}$ according to the Porter-Thomas distribution. Similarly, the probability that the resonance falls in the interval between 0 and -0.010 eV is, according to the Wigner distribution, $P_2 = 2.2 \times 10^{-4}$. Therefore we obtain a value of the cumulative probability $P = P_1 \times P_2 = 1.6 \times 10^{-7}$ which is extremely small.

## Conclusion

The results of the present experiment confirm the approximate 1/v dependence of the $^{238}U$ thermal capture cross section foreseen by most evaluations. As a consequence, the possibility of explaining the experimental values of the temperature coefficients of reactivity with a different slope of this cross section should be ruled out.


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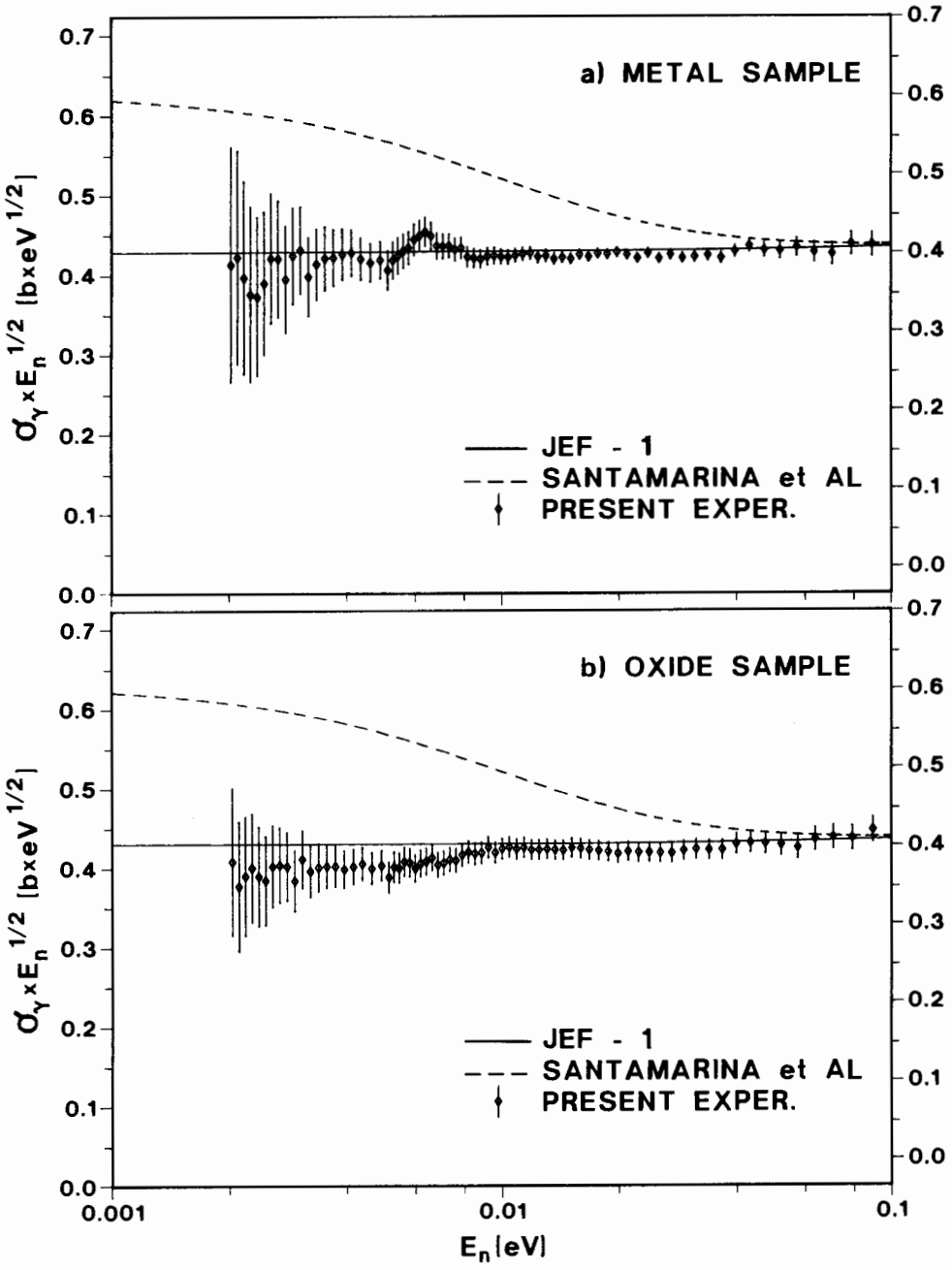


Fig. 2 Experimental points of $\sigma\gamma\sqrt{E}$ of $^{238}U$ plotted vs energy and compared to the JEF-1 and Santamarina evaluation for : a) the metal sample; b) the oxide sample. All data sets are normalized to the same value at the upper energy end.