



A.I. Shlyekher

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FUNDAMENTAL NUCLEAR CONSTANTS
USING THE OKLO NATURAL REACTOR

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ПРЯМАЯ ПРОВЕРКА ПОСТОЯНСТВА ФУНДАМЕНТАЛЬНЫХ
КОНСТАНТ ПО ДАННЫМ ОБ ЕСТЕСТВЕННОМ
ЯДЕРНОМ РЕАКТОРЕ ОКЛО

А.И.Шляхтер

А н н о т а ц и я

Показано, что положения нейтронных резонансов очень чувствительны к изменению фундаментальных ядерных констант. Из анализа измеренных изотопных сдвигов в естественном ядерном реакторе Окло получены ограничения на скорость изменения констант взаимодействий: сильных - $2 \cdot 10^{-19}$ год⁻¹, электромагнитных - $5 \cdot 10^{-18}$ год⁻¹, слабых - 10^{-12} год⁻¹. Эти пределы позволяют исключить все обсуждавшиеся в литературе варианты изменения ядерных констант.

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The positions of neutron resonances have been shown to be highly sensitive to the variation of fundamental nuclear constants. The analysis of the measured isotopic shifts in the natural fossil reactor at Oklo gives the following restrictions on the possible rates of the interaction constants variation: strong $\sim 2 \cdot 10^{-19} \text{ yr}^{-1}$, electromagnetic $\sim 5 \cdot 10^{-18} \text{ yr}^{-1}$, weak $\sim 10^{-12} \text{ yr}^{-1}$. These limits permit to exclude all the versions of nuclear constants contemporary variation discussed in the literature.

1. INTRODUCTION

The problem of the possible variation of fundamental constants is being discussed for about 40 years. Milne^{/1/} and Dirac^{/2/} were the first to notice that the constancy of physical constants during very long periods of time is a hypothesis needing experimental evidence.

Speaking on the variation of a dimensional constant one always bears in mind some "truly constant" unit of the same dimension. So any change having physical meaning can be reduced to the change of a dimensionless combination of the dimensional constants. There exist many such combinations but all of them are determined by several ones called "fundamental".

Gravitation, electromagnetism and weak interactions are determined by a single constant each (we shall denote them γ , α and β):

$$\gamma = \frac{G m^2}{\hbar c} \approx 5 \cdot 10^{-39}, \quad (1)$$

$$\alpha = \frac{e^2}{\hbar c} \approx 1/137, \quad (2)$$

$$\beta = \frac{g m^2 c}{\hbar^3} \approx 9 \cdot 10^{-6} \quad (3)$$

Here G is the Newton's constant of gravitation, g is the Fermi's weak interactions constant, m is the nucleon mass. For strong interactions it is unknown which constants pertaining to them should be regarded as fundamental. We shall estimate their variation by the relative change in the depth of nuclear potential well (see Sec.2).

Versions of the contemporary variation of the constants discussed in the literature give the relative change 10^{-10} - 10^{-12} yr⁻¹ (see refs.¹¹⁻¹⁷)

Only for α all the versions suggested had been excluded by the experimental evidence. From the data on ¹⁸⁷Re decay Dyson¹⁸ obtained (see also A.M.Wolfe et al. Phys.Rev.Lett. 32, 179 (1976).)

$$\left| \frac{1}{\alpha} \frac{d\alpha}{dt} \right| \leq 5 \cdot 10^{-15} \text{yr}^{-1} \quad (4)$$

For γ and β the experimentally established limits are about 10^{-10} yr⁻¹ and none of the proposed versions of their variation can be excluded.^{18,9}

More accurate restrictions for β are also desirable to have confidence in the correct dating of geological objects. The decay constant of β -active nucleus is proportional to β^2 so the relative change in β about 10^{-10} yr⁻¹ would give an error $\sim 2 \cdot 10^{-10} t^2$ during the time interval t . For $t \sim 10^9$ yr the error may achieve $\sim 2 \cdot 10^8$ yr.

The aim of the present paper is to draw attention to the fact that the positions of neutron resonances are very sensitive to the change of nuclear constants.

Laboratory measurements of the resonance positions during 10 years give the upper limit of the strong interaction constants variation of the same order as cosmological arguments.

On the other hand the data on the resonance positions $2 \cdot 10^9$ yr ago can be obtained from the analysis of the isotopic composition of the elements in the Oklo natural fossil reactor (see Appendix). It allows to impose the limits several orders of magni-

tude more accurate excluding all the discussed versions of nuclear constants contemporary variation by the direct evidence.

2. THE SHIFTS OF NEUTRON RESONANCES CAUSED BY THE VARIATION OF THE CONSTANTS

For the bombarding neutron a heavy nucleus represents (roughly) a potential well with the depth $V_0 \approx 50 \text{ MeV}^{10/}$. If the neutron energy is small enough ($E \lesssim 1 \text{ keV}$) the cross section exhibits narrow resonances (Fig. 1). Their positions are known to an accuracy of $\Delta_{exp} \sim 10^{-3} \text{ eV}$ (see Sec. 3). So there exist two energy scales V_0 and Δ_{exp} .

The main idea of this paper is the following. A change in V_0 by ΔV_0 would cause the shift of all the levels (including the levels of a compound nucleus i. e. neutron resonances) of the same order of magnitude. Experimental evidence for the resonance positions being constant to an accuracy of Δ_{exp} then restricts the possible relative change of V_0 by

$$\left| \frac{\Delta V_0}{V_0} \right| \leq \frac{\Delta_{exp}}{V_0} \quad (5)$$

The exact neutron - nucleus interaction can be decomposed into a sum of interaction with the average nuclear potential (that gives the single - particle motion) and residual internucleon interactions H_r . The latter cause the fragmentation of broad single - particle resonances into millions of narrow compound resonances. It is impossible to calculate the properties of a given resonance since the residual interactions are very complex^{10/}.

A change of the nuclear well depth ΔV_0 (for $H_r = \text{const}$) would lead to the uniform shift ΔE_0 of all the resonance equal to that of a single - particle resonance.

A change in residual interactions ΔH_r (for $V_0 = \text{const}$) would lead to the mutual shifts ΔE_r of the resonances which are random in magnitude and direction.

We know nothing on the relation between ΔE_0 and ΔE_r . So for a given resonance it may appear that $\Delta E_0 \approx -\Delta E_r$. But the proba-

bility of such a compensation should be small and for several resonances - negligible.

On the other hand it may appear that for some resonance $\Delta E_r \gg \Delta E_0$ resulting in its high sensitivity to the variation of the constants.

We shall assume the single - particle estimate ΔE_0 bearing in mind that Δ_{exp} obtained from the analysis of only one resonance can be relied upon only to the order of magnitude. In order to decrease the uncertainty one should analyse the shifts for a number of resonances.

Following Gamow^{/11/} we shall suppose that the strong - interaction constants variation is represented well enough by the relative change of the nuclear potential well depth $\Delta V_0/V_0$. The restriction on their possible variation is then given by (5).

The relative contribution of weak interactions to the energy of the nucleus can be estimated (to the order of magnitude) as ^{/12/} $\beta (\mu/m)^2 \approx 2 \cdot 10^{-7}$ where μ is the pion mass. From (5) we have

$$\left| \frac{\Delta \beta}{\beta} \right| \leq 5 \cdot 10^6 \frac{\Delta_{exp}}{V_0} \quad (6)$$

The variation of the electromagnetic constant α would lead to a small change in the nuclear radius R due to the variation of the Coulomb repulsion energy E_0 . We shall estimate this effect using the expression ^{/10/}

$$\frac{\Delta R}{R} = \frac{R}{KA} \left(\frac{\partial E_0}{\partial R} \right) \quad (7)$$

Here K is the nuclear compressibility coefficient ($K \approx 135$ MeV). For nuclei with $A \approx 150$ $E_0 \approx 500$ MeV, and the relative change in R is about 2.5% of the change in α . Thus the restriction on the α variation is about 20 times less accurate than that given by (5).

Summing, we can conclude that an experimental evidence for the resonance positions being constant to an accuracy of Δ_{exp} during the period of time T would impose the following restrictions on the possible rates of variation of the constants:

for the strong interactions⁻⁷

$$\left| \frac{1}{V_0} \frac{dV_0}{dt} \right| \leq 2 \cdot 10^{-8} \frac{\Delta_{exp} (eV)}{T} \quad (8)$$

for the electromagnetic

$$\left| \frac{1}{\alpha} \frac{d\alpha}{dt} \right| \leq 5 \cdot 10^{-7} \frac{\Delta_{exp} (eV)}{T} \quad (9)$$

for the weak

$$\left| \frac{1}{\beta} \frac{d\beta}{dt} \right| \leq 0.1 \frac{\Delta_{exp} (eV)}{T} \quad (10)$$

3. EVIDENCE CONCERNING THE SHIFTS OF RESONANCES

For many low - lying neutron resonances their positions had been measured to an accuracy of $\Delta_{exp} \approx 10^{-3}$ eV as long as 15 years ago^{/13/}. For example the energy of the first resonance in ¹⁵⁵Gd had been measured in 1960 its value being (0.0268 ± 0.0002) eV. The value obtained in 1969 is just the same^{/14/}. Assuming $T=10$ yr we come to the direct laboratory established evidence concerning the variation of the strong - interaction constants:

$$\left| \frac{1}{V_0} \frac{dV_0}{dt} \right| \leq 2 \cdot 10^{-12} \text{ yr}^{-1} \quad (11)$$

Similar restriction has been obtained by Davies^{/15/} using Dyson's cosmological arguments.

The resonance positions as long as $2 \cdot 10^9$ years ago can be determined using the Oklo natural reactor data (see Appendix).

The positions of the low - lying resonances determine the capture cross section for the thermal neutrons. For a single resonance at E_r having neutron and capture widths Γ_n and Γ_γ the capture cross section of neutrons with the energy E (wavelength λ) is given by the Breit - Wigner formula:

$$\sigma_c = \pi \lambda^2 g \frac{\Gamma_n \cdot \Gamma_\gamma}{(E - E_r)^2 + \left(\frac{\Gamma_n + \Gamma_\gamma}{2} \right)^2} \quad (12)$$

Here g is the statistical factor. We see that the large thermal cross section values are the most sensitive to the resonance shifts. These values are caused by the resonances lying near the thermal energy region. The variation of the thermal capture cross section of ^{149}Sm caused by the resonance shift Δ is shown on Fig.2.

The cross section value $2 \cdot 10^9$ years ago can be determined from the measured isotopic shifts at Oklo. Naudet et al.^{/16/}

measured the isotopic composition of U, Nd, and Sm in 50 samples from the reactor core. They managed to determine the fluence of thermal neutrons and the conversion coefficient C (see Appendix) from the data on U and Nd for 36 samples. In the rest ones the following diffusion of the natural elements was too large.

The cross section for ^{149}Sm can be determined from the equations (A.2) and (A.4) (see Appendix):

$$\sigma_{149} = \frac{N_{147} + N_{148}}{N_{149}} \cdot \frac{\gamma_{149}}{\gamma_{147}} \cdot \frac{e^{\sigma_{235}(1-C)\tau} - 1}{\sigma_{235}(1-C)\tau} \cdot \frac{1}{\tau} \quad (13)$$

Here σ_{149} , σ_{235} are the thermal capture cross sections of ^{149}Sm and ^{235}U , N_{147} etc. - ^{147}Sm etc. concentration, γ_{147} etc. - fission yield of ^{147}Sm etc. (they are taken from ^{/22/}).

Thus we get at the "experimental" value of ^{149}Sm capture cross section σ_{Oklo} which had been "measured" about $2 \cdot 10^9$ years ago:

$$\sigma_{Oklo} = (55 \pm 8) \cdot 10^3 \text{ barn.}$$

Taking into account two standard errors we come to the following restriction on the possible shift of the first resonance in ^{149}Sm (see Fig.2):

$$|\Delta_{exp}| \lesssim 20 \cdot 10^{-3} \text{ eV.} \quad (15)$$

Such a small shift would not change the capture cross sections of Nd and U isotopes. So the direct use of the neutron flu-

ences from ^{/16/} is correct. - 9 -

Table 1 is a list of the upper limits obtained from (15) and (8-10) assuming $T = 2 \cdot 10^9$ years. For comparison the earlier limits by Dyson^{/8/} and Davies^{/15/} are also listed below.

Table 1

Relative change years ⁻¹	Dyson, Davies	Present work
$\left \frac{1}{V_0} \frac{dV_0}{dt} \right $	$2 \cdot 10^{-12}$	$2 \cdot 10^{-19}$
$\left \frac{1}{\alpha} \frac{d\alpha}{dt} \right $	$5 \cdot 10^{-15}$	$5 \cdot 10^{-18}$
$\left \frac{1}{\beta} \frac{d\beta}{dt} \right $	10^{-10}	10^{-12}

It is interesting to observe that the rate of the variation $\geq 10^3$ times exceeding these limits would cause a drastic change of many capture cross section values during $\sim 10^9$ years. On the one hand it would make the reliable interpretation of the isotopic shifts at Oklo almost impossible. On the other hand ^{235}U would appear to be a strongly capturing nucleus during some period of time. It would turn the spontaneous chain reaction in the uranium deposits into a usual occurrence. The isotopic composition of natural uranium would then vary considerably from place to place.

Thus the fact that the ^{235}U abundance is remarkably uniform everywhere (except Oklo) is one more argument in the favor of the constancy of the fundamental nuclear constants.

4. WHAT IS INTERESTING TO MEASURE AT OKLO ?

The results of Sec. 3 permit to exclude all the versions of the nuclear constants contemporary variation discussed in the literature. But the constants may appear to vary in some unpredictable way^{/8/}. So it is an important task to obtain as precise and reliable restrictions as it is possible.

We have seen in Sec. 3 that the large capture cross section values are the most sensitive to the shifts of neutron resonances. Unfortunately the equilibrium concentration of the strong capture cross sections at Oklo is exceedingly low. (see Appendix). The following diffusion and the change of isotopes between different elements cause the change in isotopic composition of the same order.

For this reason it is desirable to perform the accurate measurements of the isotopic composition of all rare earth fission products (in a given sample). The data obtained would allow to determine Δ_{exp} for several resonances thus raising the reliability of the restrictions. The existing data are scarce and insufficiently accurate giving Δ_{exp} far exceeding (15).

It should be emphasized that apart from the assumptions made in Sec. 2 the very fact of the neutron resonance shift really occurring would appear to be an important discovery. It would show that some fundamental constants do really vary with time thus compelling a reconsideration of the basic physical theories.

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APPENDIX. THE OKLO NATURAL FOSSIL REACTOR

We shall briefly discuss here some facts concerning the "Oklo phenomenon". The majority of the review and original papers is contained in refs.^{/16,17/}.

The natural uranium consists of two isotopes: ^{238}U ($T_{1/2} = 4.51 \cdot 10^9 \text{yr}$) and ^{235}U ($T_{1/2} = 0.71 \cdot 10^9 \text{yr}$). Only the latter undergoes fission being irradiated by thermal neutrons. The contemporary enrichment of natural uranium by ^{235}U is too low for the spontaneous chain reaction occurring in uranium deposits.

But in the remote past the natural abundance of ^{235}U was greater (Fig. 3, curve I). Two billion years ago it exceeded 3% being the same as in modern power producing reactors.

The chain reaction could start spontaneously if the conditions were favourable /18,19/. As the result the ^{235}U enrichment in the deposit would become lower than everywhere. But the search for an anomaly of ^{235}U enrichment in uranium deposits brought no result for a long time.

It was only in 1972 that the abnormal isotopic composition of uranium had been found in the deposit at Oklo (Gabon)^{/20/}. The average enrichment was about 0.6% in 500 tons of uranium ore and in several samples — lower than 0.3%.

The systematic study of this phenomenon had been carried out within a specially established "Project Franceville" headed by R. Naudet (French Atomic Energy Commission).

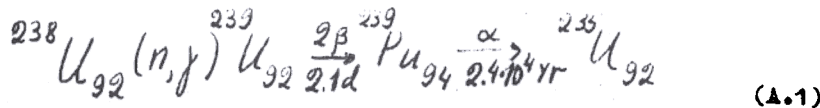
It appeared that about $2 \cdot 10^9$ years ago several natural reactors had been acting at Oklo deposit (Fig. 4). The total energy released equals to about 10^{11} kW.h, the neutron fluence at certain points exceeded $1.5 \cdot 10^{21} \text{n/cm}^2$.

The fact of spontaneous chain reaction having occurred at Oklo is now considered to be proved.

Let us reveal briefly the simplest methods used to estimate the parameters essential for the present work.

Duration of the reactions is determined supposing that their rate was constant. ^{235}U is formed from ^{238}U according to the

Following reactions:



If the duration was less than 10^4 years then about 50% of fissions must be due to ${}^{239}\text{Pu}$. This fraction can be estimated from the measured isotopic composition of fission products. The data on the rare earth elements suggest that they are almost completely the products of ${}^{235}\text{U}$ thermal fission. ${}^{239}\text{Pu}$ contribution is about 2% leading to the estimate of the duration of the order of $5 \cdot 10^5$ years.

The age of the Oklo phenomenon T has been determined by several independent methods giving consistent results. The simplest of them is based on the measurement of ${}^{235}\text{U}$ enrichment and the ratio "number of fissions/total number of U atoms" f/U_T . Before the reaction had begun the ${}^{235}\text{U}$ abundance at Oklo had been normal and varied with time respecting the curve I on Fig. 3.

The duration was small in comparison with the ${}^{235}\text{U}$ half life period. So the AB line representing the reactions is vertical. After the reaction had finished ${}^{235}\text{U}$ enrichment had dropped and thereafter changed according to the curve II. The value of the (f/U_T) ratio gives the AB length. For example in the sample 2'P-1181 f/U_T is about 3.4% and ${}^{235}\text{U}$ enrichment - 0.3% / 21. Fig. 3 shows that 3% of uranium could not burn up later than $2 \cdot 10^9$ years ago. More accurate considerations (taking into account the ${}^{235}\text{U}$ formation according to the reactions (A.1)) give $T = (1.7 \pm 2.0) \cdot 10^9$ years.

The fluence of thermal neutrons \bar{c} can be determined from the change in the isotopic composition of the elements in the reactor core. The strongly capturing nuclei reach the equilibrium concentration quickly. One can estimate \bar{c} considering the Sm isotopic composition. ${}^{147}\text{Sm}$ is accumulated in the course of reaction partially converting into ${}^{148}\text{Sm}$. The latter has very small fission yield and capture cross section. Considering the ratio $(N_{147} + N_{148})/N_{149}$ (N_{147} is the ${}^{147}\text{Sm}$ concentration etc.)

we are led to the equation:

$$\frac{N_{147} + N_{148}}{N_{149}} = \frac{\gamma_{147}}{\gamma_{149}} \cdot \frac{N_{235}}{N_{235}} \cdot \bar{c}_{149} \bar{c} \quad (\text{A.2})$$

Here γ_{147} etc. is the fission yield of ${}^{147}\text{Sm}$ etc., N_{235}/N_{235} is the ratio of the average ${}^{235}\text{U}$ concentration during the period of the reaction to its final concentration. \bar{c}_{149} is the ${}^{149}\text{Sm}$ capture cross section. ($\bar{c}_{149} \approx 60 \cdot 10^3$ barn). For $\bar{c} > 10^{20} \text{ n/cm}^2$ ${}^{149}\text{Sm}$ equilibrium concentration should be very small. Indeed the average value of the ratio $(N_{147} + N_{148})/N_{149}$ exceeds 100 while for the fission products and natural Sm it is about 2.13, 22/

Extremely small concentration of the strongly capturing nuclei at Oklo is the most convincing proof of the thermal neutron chain reaction really having occurred at Oklo.

The fluence \bar{c} can also be determined from the analysis of the Nd isotopic composition. For example the ${}^{143}\text{Nd}$ concentration can be described by the system:

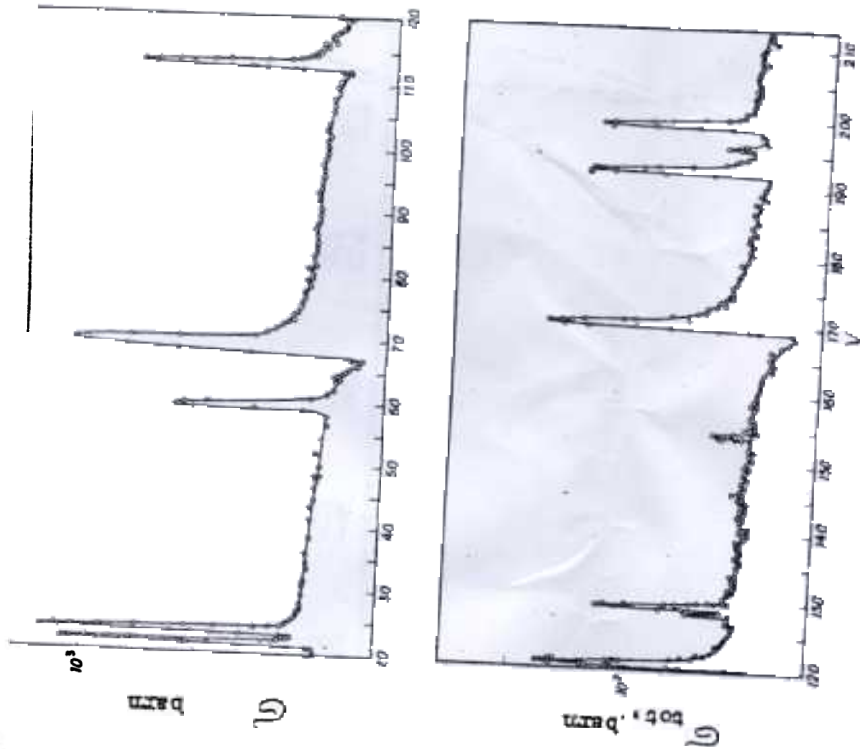
$$dN_{143} = (\gamma_{143} \bar{c}_{235} N_{235} - \bar{c}_{143} N_{143}) dt, \quad (\text{A.3})$$

$$dN_{235} = -\bar{c}_{235} (1 - C) N_{235} dt. \quad (\text{A.4})$$

Here \bar{c}_{235} is the ${}^{235}\text{U}$ fission cross section, C is the "conversion coefficient" - the fraction of ${}^{235}\text{U}$ formed by the reactions (A.1)

Adopting several simplifying assumptions (giving an uncertainty about 20%) one can determine \bar{c} and C from the measured isotopic composition of U and Nd.

The fluences determined in such a way are about 10^{21} n/cm^2 . They vary smoothly across the reactor core showing that the mutual displacement of the different parts of the reaction zone during $2 \cdot 10^9$ years was small.



The energy dependence of the neutron capture cross section of ^{232}Th .

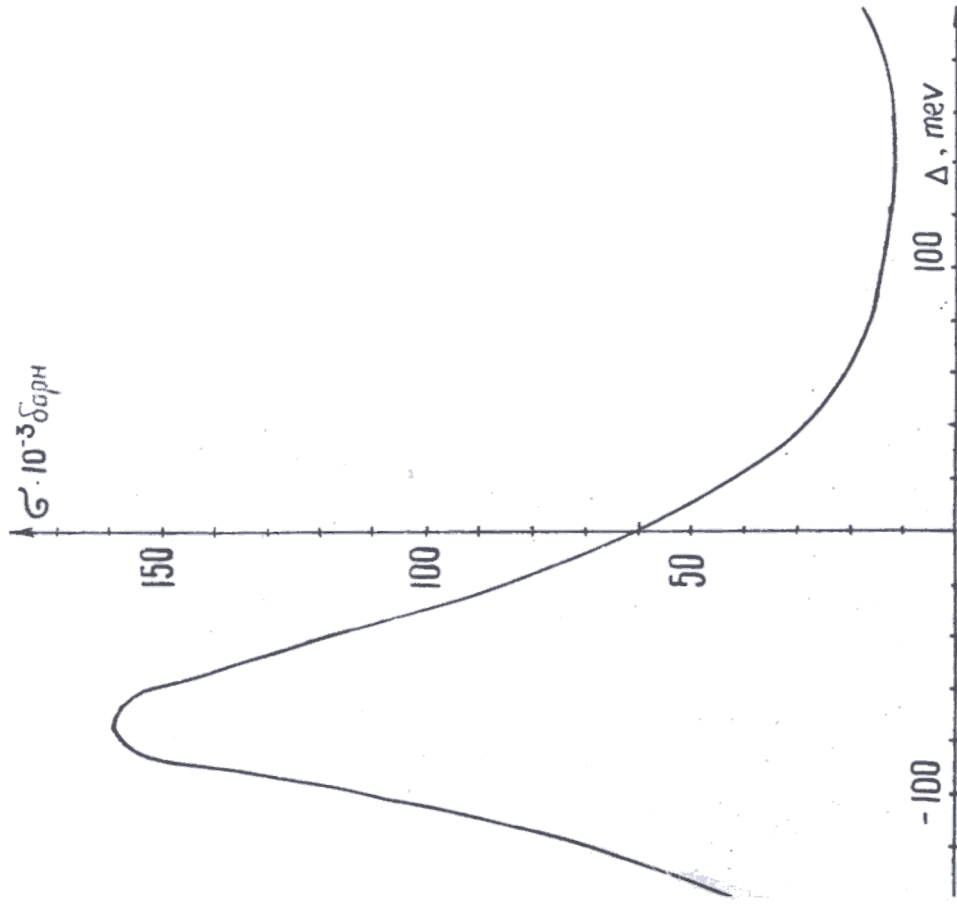


Fig. The energy dependence of the thermal neutron capture cross section of ^{149}Sm caused by the shift of all resonances (with parameters $\Gamma = 0.025\text{eV}$).

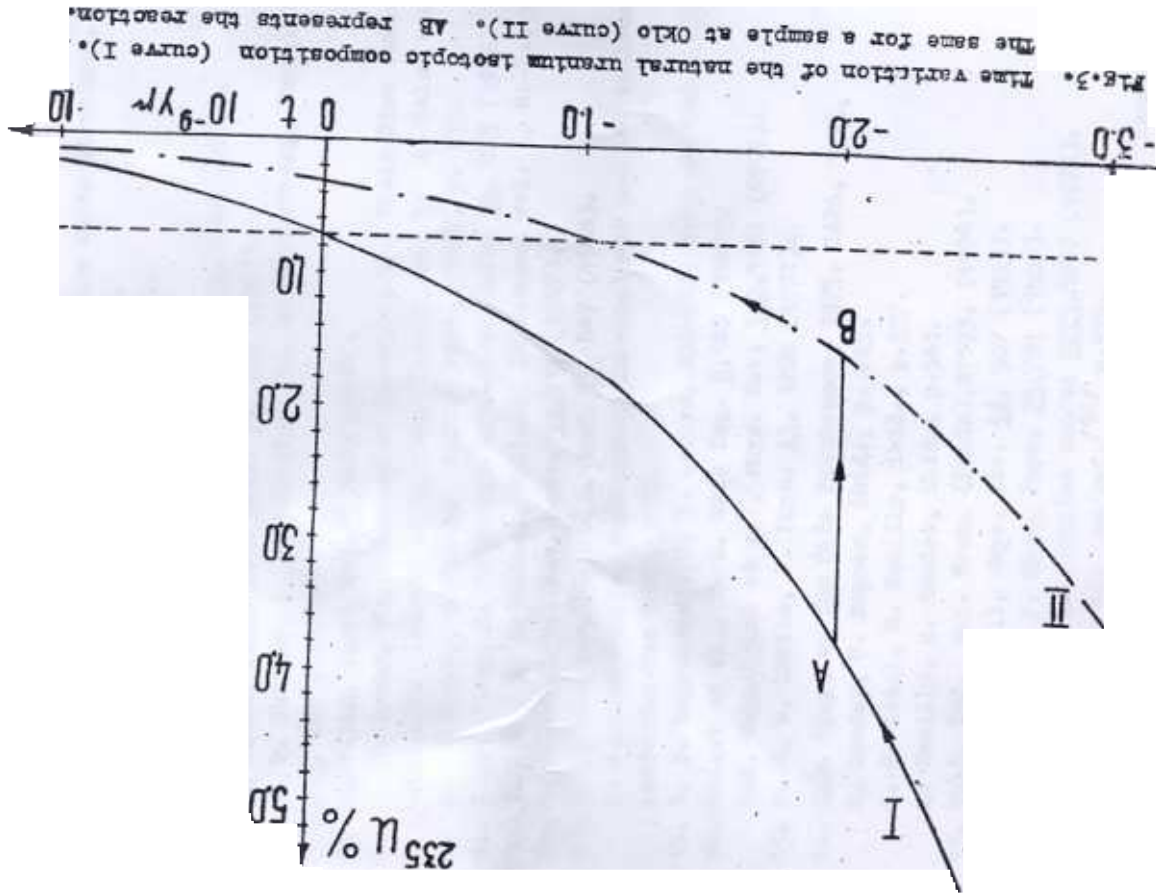


FIG. 3. Time variation of the natural uranium isotopic composition (curve I). The same for a sample at Oklo (curve II). AB represents the reaction.

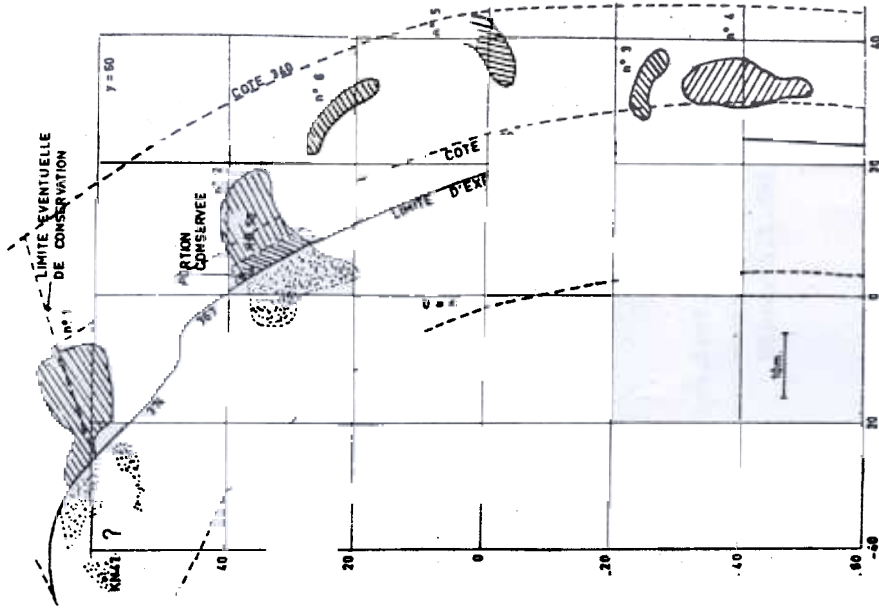


Fig. 4. Six reaction zones known in 1975. Zones N°1 and N°2 had been partially exploited before the discovery of the anomaly.

References

1. E. A. Milne, Relativity, Gravitation and World Structure, Oxford, Clarendon Press, 1935, p. 292.
2. P. A. M. Dirac, Nature 139, 323 (1937).
3. E. Teller, Phys. Rev. 73, 801 (1948).
4. G. Rosen, Canadian Journ. of Physics 45, 2383 (1967).
5. G. Gamow, Phys. Rev. Lett. 19, 759 (1967).
6. V. P. Chechev, L. E. Gurevich, Ya. M. Kramarovsky, Phys.Lett. B42, 261 (1972).
7. S. Malin, Phys. Rev.D9,3228 (1974).
8. F. J. Dyson, The Fundamental Constants and Their Time Variation. In Aspects of Quantum Theory, ed. by A. Salam and E. P. Wigner, Cambridge Univ. Press, 1972, p. 213.
9. C.-w. Cheng, R. Stothers, Phys. Rev. Lett. 36, 833 (1976).
10. A. Bohr, B. Mottelson, Nuclear Structure, vol. I, N.-Y., 1969.
11. G. Gamow, Phys. Rev. Lett. 19, 913 (1967).
12. I. S. Shapiro, Usp. Fiz. Nauk 95, 647 (1968).
R. J. Blin-Stoyle, Fundamental Interactions and the Nucleus, Amsterdam-New York, 1973.
13. S. F. Mughabghab, D. I. Garber, BNL-325, 3-rd ed. vol.I (1973)
14. Moller et al., Nucl. Sci. Eng. 8, 183 (1960).
S.F. Mughabghab et al., Phys. Rev. 180, 1131 (1969).
15. P. C. W. Davies, J. Phys. A5, 1296 (1972).
16. Int. Symp. on the Oklo Phenomenon, 1975, IAEA, 1975.
R. Naudet, G. Renson, *ibid.*, p. 265.
J.-F. Dozol, M. Neuilly, *ibid.*, p.357.
M. Neuilly, R. Naudet, *ibid.*, p.541.
17. Bull. Inf. Sci. Tech. (Paris), N°193, (1974).
18. G. F. Wetherill, Phys. Rev. 92, 907 (1953).
19. P. K. Kuroda, J. Chem. Phys. 25, 781 (1956).
20. M. Neuilly et al. Comptes rendus D275, 1847 (1972).
21. W. J. Maack et al. in ref. ^{/16/} p.319.
22. M. E. Meek, B. F. Rider, Compilation of Fission Product Yields, Vallecitos Nuclear Center (1974).

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Editor A.I.Shlyakhter