

Natural nuclear reactor at Oklo and variation of fundamental constants: Computation of neutronics of a fresh core

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Using modern methods of reactor physics, we performed full-scale calculations of the Oklo natural reactor. For reliability, we used recent versions of two Monte Carlo codes: the Russian code MCU-REA and the well-known international code MCNP. Both codes produced similar results. We constructed a computer model of the Oklo reactor zone *RZ2* which takes into account all details of design and composition. The calculations were performed for three fresh cores with different uranium contents. Multiplication factors, reactivities, and neutron fluxes were calculated. We also estimated the temperature and void effects for the fresh core. As would be expected, we found for the fresh core a significant difference between reactor and Maxwell spectra, which had been used before for averaging cross sections in the Oklo reactor. The averaged cross section of $^{149}_{62}\text{Sm}$ and its dependence on the shift of a resonance position E_r (due to variation of fundamental constants) are significantly different from previous results. Contrary to the results of previous papers, we found no evidence of a change of the samarium cross section: a possible shift of the resonance energy is given by the limits $-73 \leq \Delta E_r \leq 62$ meV. Following tradition, we have used formulas of Damour and Dyson to estimate the rate of change of the fine structure constant α . We obtain new, more accurate limits of $-4 \times 10^{-17} \leq \dot{\alpha}/\alpha \leq 3 \times 10^{-17} \text{ yr}^{-1}$. Further improvement of the accuracy of the limits can be achieved by taking account of the core burn-up. These calculations are in progress.

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I. INTRODUCTION

The discovery of the Oklo natural nuclear reactor in Gabon (West Africa) in 1972 was possibly one of the most momentous events in reactor physics since 1942, when Enrico Fermi and his team achieved an artificial self-sustained fission chain reaction. Soon after the discovery of the ancient natural reactor [1–3], one the authors of the present paper (Yu.P.) and his postgraduate student A. I. Shlyakhter realized that the Oklo phenomenon could be used to find precise limits on possible changes of fundamental constants. At that time, they considered probabilistic predictions of unknown absorption cross sections based on static nuclear properties. Near the neutron binding energy ($B_n = 6\text{--}8$ MeV) the resonances form a fence with a mean separation of tens of electron volts. The magnitude of the cross section depends on the proximity of the energy E_{th} of the thermal neutron to the nearest resonance. If the energy $E_{\text{th}} = 25$ meV falls directly on a resonance, then the cross section increases to as much $10^5\text{--}10^6$ b [4–6]. If the cross sections have changed with time, then the entire fence of resonances as a whole has shifted by a small amount ΔE_r . This shift can be established most accurately from the change of the cross section of strong absorbers (for instance, $^{149}_{62}\text{Sm}$). The estimate of ΔE_r on the basis of experimental data for the Oklo reactor ($\Delta E_r \leq 3 \times 10^{-17}$ eV/yr) [7–10] allowed them to estimate a possible limit of the rate of change of fundamental constants. This estimate has remained the most accurate one for 20 years. In 1996, Damour and Dyson checked and confirmed the results of Refs. [7–9]; they were the first to calculate the dependence of the capture cross section on

the temperature T_C of the core: $\hat{\sigma}_{r,\text{Sm}}(T_C)$ [11]. In 2000, Fujii *et al.* significantly reduced the experimental error of the cross section $\hat{\sigma}_{r,\text{Sm}}(T_C)$ [12]. In both papers, the authors averaged the samarium cross section with a Maxwell velocity spectrum over a wide interval of core temperature T_C .

After the publication of Ref. [11], one of the authors of the present paper (Yu.P.) realized that the limit on the change of the cross section can be significantly improved at least in two directions:

- (i) Instead of a Maxwell distribution, the samarium cross section should be averaged with the spectrum of the Oklo reactor which contains the tail of the Fermi spectrum of slowing down epithermal neutrons. This question was also discussed by Lamoreaux and Torgerson [13].
- (ii) The range of admissible core temperatures T_C can be significantly reduced, assuming that T_C is the equilibrium temperature at which the effective multiplication factor K_{eff} of the reactor is equal to unity. On account of the negative power coefficient (void + temperature), such a reactor state will be maintained for a long time until the burn-up results in a reduction of the reactivity excess and hence of T_C . Since $^{149}_{62}\text{Sm}$ burns up about 100 times faster than $^{235}_{92}\text{U}$, the core will contain only that amount of samarium that was generated immediately before the reactor shut down. Therefore, one needs to know T_C at the end of the cycle.

To solve this problem, one must use modern neutron-physical and thermohydrodynamical methods of reactor calculations. We have built a complete computer model of the Oklo reactor core *RZ2* and established its material composition. We chose three variants of its initial composition in order to estimate its effect on the spread of results. To increase

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the reliability of the results, we used modern versions of two Monte Carlo codes. One of them, developed at the Kurchatov Institute, is the licensed Russian code MCU-REA with the library DLC/MCUDAT-2.2 of nuclear data [14]; the other is the well-known international code MCNP4C with library ENDF/B-VI [15]. Both codes give similar results. We have calculated the multiplication factors, reactivity, and neutron flux for the fresh cores, and the void and temperature effects [16]. As expected, the reactor spectrum differs strongly from a Maxwell distribution (see below). The cross section $\hat{\sigma}_{r,Sm}(T_C, \Delta E_r)$, averaged with this distribution, is significantly different from the cross section averaged with a Maxwell distribution [17]. We use our result for the averaged cross section to estimate the position of resonances at the time of Oklo reactor activity. This allows us to obtain the most accurate limits on the change of the fine structure constant in the past.

The paper is organized as follows. In Sec. II, we describe briefly the history of the discovery of the natural Oklo reactor and itemize the main parameters of its cores. We consider mainly the RZ2 core. We describe in detail the neutronics of this core calculated by modern Monte Carlo codes. However, simple semianalytical considerations are also useful to clarify the picture. We consider the power effect, which is the sum of the temperature and void effects. At the end of the section, we discuss the computational difficulties in the calculations of the unusually large RZ2 core and demonstrate that Monte Carlo methods are, in general, inadequate for the calculations of core burn-up.

The main result of Sec. II is the neutron spectrum in the fresh core. In Sec. III, we apply this spectrum to obtain the averaged cross section of $^{149}_{62}\text{Sm}$ in the past. We begin this section with an explanation of the way to obtain precise limits on the variation of fundamental constants using the available Oklo reactor data. We describe different approaches to the problem and relate the variation of the constants to the change in the averaged cross sections for thermal neutrons. Using our value for the cross section of $^{149}_{62}\text{Sm}$ we obtain limits on the variation of the fine structure constant which is the best available at the moment. At the end of this section, we compare our result with the results obtained in other papers and discuss possible reasons for differences.

II. NEUTRONICS OF THE FRESH CORE

A. History of the discovery and parameters of the Oklo reactor

1. History of the discovery of the natural reactor

The first physicist to say that a nuclear chain reaction could have been more easily realized a billion years ago was Yakov Borisovich Zeldovich in 1941 [18]. At that time, he was considering the possibility of producing a fission chain reaction in a homogeneous mixture of natural uranium with ordinary water. His calculations (with Yu. B. Khariton) showed that this could be achieved with an approximately two-fold enrichment of natural uranium [19,20]. A billion years ago the relative concentration of the light uranium isotope was significantly higher, and a chain reaction was possible in a mixture of natural uranium and water. “Yakov Borisovich said nothing about the possibility of a natural reactor, but his thoughts directly

led us to the natural reactor discovered in Gabon in 1972,” reminisced I. I. Gurevich [18]. Later, in 1957, G. Whetherill and M. Inghram arrived at the same conclusion [21,22]. Going from the present concentration of uranium in pitchblende, they concluded that about 2 billion years ago, when the proportion of $^{235}_{92}\text{U}$ exceeded 3%, conditions could have been close to critical. Three years later, P. Kuroda [23,24] showed that if in the distant past, water had been present in such deposits, then the neutron multiplication factor (K_∞) for an infinite medium could have exceeded unity and a spontaneous chain reaction could have arisen. But before 1972, no trace of a natural reactor had been found. On June 7, 1972, during a routine mass-spectroscopic analysis in the French Pierrelatte factory that produced enriched fuel, H. Bouzigues [1,3] noticed that the initial uranium hexafluoride contained $\zeta_5 = 0.717\%$ of $^{235}_{92}\text{U}$ atoms instead of the 0.720%, which is the usual concentration in terrestrial rock, meteorites, and lunar samples. The French Atomic Energy Authority (CEA) began an investigation into this anomaly, which was called the Oklo phenomenon. The results of this research were published in the proceedings of two IAEA symposia [2,25]. The simplest hypothesis of a contamination of the uranium by depleted tails of the separation process was checked and shown to be wrong. Over a large number of steps of the production process, the anomaly was traced to the Munana factory near Franceville (Gabon) where the ore was enriched. The ore with a mean uranium concentration of (0.4–0.5)% had been delivered from the Oklo deposit. The isotope analysis of the uranium-rich samples showed a significant depletion of the $^{235}_{92}\text{U}$ isotope and also a departure from the natural distribution of those rare earth isotopes known as fission products [1,3,26,27]. This served as proof of the existence in the distant past of a spontaneous chain reaction. It had taken less than 3 months to produce this proof. A retrospective analysis of documents and samples of the Munana enrichment factory showed that in 1970–72 ore was delivered for processing that contained at times up to 20% of uranium depleted to 0.64% of isotope $^{235}_{92}\text{U}$ [28]. Considering that the ore was mixed during mining, the uranium concentration could have been even higher in some samples, and the depletion even stronger. Altogether more than 700 tons of depleted uranium has been mined that took part in the chain reaction. The deficit of $^{235}_{92}\text{U}$ (which had not been noticed at first) was about 200 kg. By agreement with the government of Gabon, the uranium ore production company of Franceville (COMUF) agreed to halt mining in the region of the natural reactor. A Franco-Gabon group headed by R. Naudet began a systematic study of the Oklo phenomenon. Numerous samples, obtained by boring, were sent for analysis to various laboratories around the world. Their results allowed a reconstruction of the functioning of the reactor in the Precambrian epoch.

2. Geological history of the Oklo deposit

As was shown by U/Pb analysis, the Oklo deposit with a uranium concentration of about 0.5% in the sediment layer was formed about 2×10^9 years ago [29–31]. During this epoch, an important biological process was taking place: the transition from prokaryotes, i.e., cells without a nucleus, to

eucaryotes—more complex unicellular forms containing a nucleus. The eucaryotes began to absorb carbon oxide, and hence they saturated the atmosphere with oxygen. Under the influence of oxygen, the uranium oxides began transforming into forms containing more oxygen, which are soluble in water. Rains washed them into an ancient river, forming at its mouth a sandstone sediment, rich in uranium, 4 to 10 m thick and 600 to 900 m wide [31]. The heavier uranium particles settled more quickly to the bottom of the nearly stagnant water of the river delta. As a result, the sandstone layer became enriched with uranium up to 0.5% (as in an enrichment factory). After its formation, the uranium-rich layer, which was resting on a basalt bed, was covered by sediments and sank to a depth of 4 km. The pressure on this layer was 100 MPa [32], which fractured the layer and allowed groundwater to enter the clefts. Under the action of the filtered water, which was subjected to high pressure, and as a result of not completely understood processes, lenses formed with very high uranium concentrations (up to 20–60% in the ore) with widths of 10–20 m and thicknesses of about 1 m [33]. The chain reaction took place in these lenses. After the end of the chain reaction, the deposit was raised to the surface by complicated tectonic processes and became accessible for mining. Within tens of meters, six reaction centers were found immediately, and altogether the remains of 17 cores were found [34].

The age T_0 of the reactor was determined from the total number of ^{235}U nuclei burned up in the past, $N_{5b}(d)$, and the number of nuclei existing today, $N_5(T_0)$ (here N_5 is the density of ^{235}U and d is the duration of the chain reaction). To determine T_0 in this way, it is necessary to know the number of ^{239}Pu nuclei formed as a result of neutron capture by ^{238}U and decayed to ^{235}U , and the fluence $\Psi = \Phi d$ (Φ being the neutron flux). Another independent method consists of determining the amount of lead formed as a result of the decay of ^{235}U , assuming that it did not occur in such a quantity in the initial deposit [29]. Both methods yield $T_0 = 1.81(5) \times 10^9$ yr [10, 35]. Below we assume in our calculations the value of $T_0 = 1.8 \times 10^9$ yr.

The duration of the work of the reactor can be established from the amount of ^{239}Pu formed. One can separate the decayed ^{239}Pu from the decayed ^{235}U using the different relative yields of Nd isotopes: $\delta_{\text{Nd}}^9 = {}^{150}\text{Nd}/({}^{143}\text{Nd} + {}^{144}\text{Nd}) = 0.1175$ for ^{239}Pu , and $\delta_{\text{Nd}}^9 = 0.0566$ for ^{235}U [36]. However, this comparison is masked by the fission of ^{238}U by fast neutrons: $\delta_{\text{Nd}}^8 = 0.1336$. Taking into account this contribution, one arrives at an estimate of $d \sim 0.6$ million yr [37]. This was the value we adopted in our calculations.

The total energy yield of the reactor has been estimated to be 1.5×10^4 MWa (MW-years) [38]. Such a fission energy is produced by two blocks of the Leningrad atomic power station with 100% load in 2.3 yr. Assuming a mean duration of $d = 6 \times 10^5$ yr for the work of the reactor, one gets a mean power output of only $P_p = 25$ kW.

B. Composition and size of the Oklo RZ2 reactor

The cores of the Oklo reactor have been numbered. The most complete data are available for core RZ2. This core of

the Oklo reactor is of the shape of an irregular rectangular plate that lies on a basalt bed at an angle of 45° . The thickness of the plate $H = 1$ m, its width $b = 11\text{--}12$ m, and its length $l = 19\text{--}20$ m (see Fig.8a in Refs. [38] and [29]). Thus, the volume of the RZ2 core is about 240 m^3 . Since in the case of large longitudinal and transverse sizes the shape of the reactor is not essential, we have assumed as a reactor model a flat cylinder of height $H = 1$ m and radius R which is determined by the core burn-up. The energy yield is $P_p d = 1.5 \times 10^4 \text{ MWa} \simeq 5.48 \times 10^6 \text{ MWd}$. At a consumption of ^{235}U of $g = 1.3 \text{ g/MWd}$ (MW-days) [40], the total amount of burned-up fissile matter is

$$\Delta M_b = g P_p d = 7.12 \text{ tons.} \tag{1}$$

Taking into account that half of the burned ^{235}U isotope is replenished from the decay of the produced ^{239}Pu , we find the original mass of the burned-up ^{235}U to be

$$\Delta M_5 = 4.75 \text{ tons.} \tag{2}$$

In the case of a uniform burn-up, the average density of the burned-up ^{235}U is

$$\Delta \gamma_5(d) = \frac{\Delta M_5}{\pi R^2 H} = \frac{1.51}{R^2} \text{ g/cm}^3, \tag{3}$$

where R is given in meters. The relative average initial burn-up is

$$\bar{\gamma}_5(d) = \frac{\Delta \gamma_5(d)}{\gamma_{5,i}(0)} = \frac{1.51 \text{ g/cm}^3}{\gamma_{5,i}(0) R_i^2} \text{ and } R_i = \left[\frac{1.51 \text{ g/cm}^3}{\gamma_{5,i}(0) \bar{\gamma}_5(d)} \right]^{1/2}. \tag{4}$$

Processing the data of Table 2 from Ref. [38] gives a value of $\bar{\gamma}_5(T_0) \simeq 50\%$ for the present-day average over the core. In the past, it was 1.355 times smaller (see below) on account of the higher concentration $\gamma_{5,i}(0)$ of uranium, i.e., $\bar{\gamma}_5(0) = 36.9\%$. Thus, the radius is given by the formula

$$R_i = \left[\frac{4.09 \text{ g/cm}^3}{\gamma_{5,i}(0)} \right]^{1/2} \text{ m.} \tag{5}$$

The approximate composition of the rock in core RZ2 is shown in Table 1 of Ref. [10]. On the basis of these data, one can calculate the elemental composition of the ore by weight (see the penultimate column of Table I). For comparison, we show in the last column the composition by weight from the

TABLE I. Present-day composition of the empty rock [10].

Chemical	% by composition	Elemental composition	Atomic weight, A_i	% by weight	% by weight [41]
SiO ₂	43.00	O	15.999	44.04	44
Al ₂ O ₃	25.73	Si	28.086	20.10	20
FeO	14.53	Al	26.982	13.62	16
Fe ₂ O ₃	4.47	Fe	55.847	14.42	11
MgO	10.43	Mg	24.305	6.30	4
K ₂ O	1.84	K	39.098	1.53	2
Sum	100			100	97

TABLE II. Present-day and initial composition of the ore in core *RZ2* [44].

	<i>i</i>	1	2	3
Present-day fraction of U in the dry ore (%)	$Y_{U,i}(T_0)$	35	45	55
Present-day density of the dry ore (g/cm ³) (Fig. 1)	$\gamma_i(T_0)$	2.84	3.29	3.82
Present-day density of U in the dry ore (g/cm ³)	$\gamma_{U,i}(T_0)$	0.994	1.481	2.101
Present-day density of UO ₂ in the dry ore (g/cm ³)	$\gamma_{UO_2,i}(T_0)$	1.128	1.680	2.38
Present-day density of the dry rock with Pb (g/cm ³)	$\gamma_{0,i}(T_0)$	1.71	1.61	1.44
Density of H ₂ O at $T = 300$ K (with water of crystallization)	γ_{H_2O}	0.355	0.355	0.355
Initial density of $^{238}_{92}\text{U}$ (g/cm ³)	$\gamma_{8,i}(0)$	1.305	1.952	2.758
Initial density of $^{235}_{92}\text{U}$ (g/cm ³)	$\gamma_{5,i}(0)$	4.216×10^{-2}	6.28×10^{-2}	8.91×10^{-2}
Initial density of U (g/cm ³)	$\gamma_{U,i}(0)$	1.347	2.015	2.847
Initial density of UO ₂ (g/cm ³)	$\gamma_{UO_2,i}(0)$	1.528	2.286	3.230
Initial density of ore (g/cm ³)	$\gamma_i(0)$	3.51	4.08	4.78
Initial density of ore without UO ₂ (g/cm ³)	$\gamma_{0,i}(0)$	1.98	1.80	1.55
Initial fraction of U in the dry ore (%)	$Y_{U,i}(0)$	38.4	49.3	59.6

book of Yu. A. Shukolyukov (Table 2.1 of Ref. [41]), which was based on early data of R. Naudet [42]. These values coincide within 10%. Since all those elements are relatively weakly absorbing, such differences do not play any practical role.

It is much more important to know the amounts of uranium and water present at the beginning of the work of the reactor. The connection between the uranium content in the core (in vol.%) and the density of the dehydrated core has been measured experimentally in Ref. [43] (Fig. 1). The content of

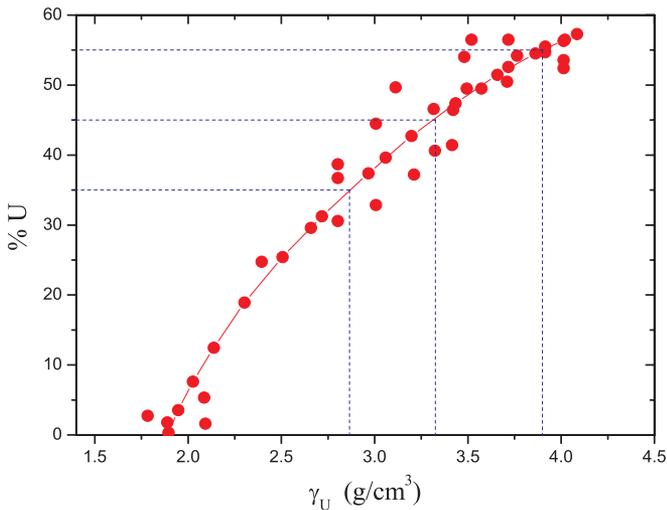


FIG. 1. (Color online) Present-day abundance of uranium (in volume percent of dry ore) in dependence on the dry ore density γ_U in the Oklo reactor [43]. Three initial variants of the $Y_{U,i}(T_0) = 35, 45, 55\%$ U in dry ore are shown.

uranium in the core varies greatly between different samples. To determine the influence of the uranium content on the reactor parameters, we have chosen three initial values for the density of uranium in the dehydrated ore: $Y_{U,i}(T_0) = 35, 45,$ and 55% , taken to be constant over the reactor. The value of the ore density $\gamma_i(T_0)$ that corresponds to $Y_{U,i}(T_0)$ is shown in the second row of Table II (see Fig. 1). In the fifth row of Table II, we show the density of the empty rock. The density of water in the reactor is $0.3\text{--}0.5$ g/cm³ [34]. This water consists of bound (crystalline) and unbound water which evaporates after 100°C . In our reactor model, we assumed a total density of water of $\gamma_{H_2O} = 0.355$ g/cm³, of which 0.155 g/cm³ was taken for the density of unbound water. Assuming a porosity of about 6%, one can take for the density of dry ore with water the same value as for the dehydrated ore.

The density of $^{238}_{92}\text{U}$ of the fresh core *RZ2* at the epoch of the formation of the reactor was

$$\gamma_8(0) = \gamma_8(T_0)(1 - \zeta_5) \exp(+T_0/\tau_8), \quad (6)$$

where the lifetime of $^{238}_{92}\text{U}$ is $\tau_8 = 6.45 \times 10^9$ yr. The value of $\gamma_U^8(0)$ increases on account of the decay of uranium into lead. The density of $^{235}_{92}\text{U}$ [$\gamma_{U,5}(0)$, g/cm³] in the fresh core ($\tau_5 = 1.015 \times 10^9$ yr) is

$$\gamma_{U,5}(0) = \gamma_{U,5}(T_0)\zeta_5 \exp(+T_0/\tau_5). \quad (7)$$

The values of $\gamma_{5,i}(0)$ and $\gamma_{8,i}(0)$ are shown in Table II. Also in the table are the calculated values of the densities of uranium $\gamma_{U,i}(0)$ and of the empty rock (without Pb), and the new fraction of uranium in the dry ore at the beginning of the cycle of the Oklo reactor. From Eq. (6) and Eq. (7) we get the ratio

$$\begin{aligned} \gamma_{U,i}(0)/\gamma_{U,i}(T_0) &= (1 - \zeta_5) \exp(T_0/\tau_8) \\ &+ \zeta_5 \exp(T_0/\tau_5) = 1.355, \end{aligned} \quad (8)$$

TABLE III. Specific weight $\gamma_{k,i}(0)$ and nuclear density $N_{k,i}(0)$ in the compositions of three variants of the reactor core [44].

k	Elemental composition	i $Y_{U,i}(0)$, %	1 38.4		2 49.4		3 59.6		
			Atomic weight, A_k (g/mol)	$\gamma_{k,1}(0)$	$N_{k,1}(0)$	$\gamma_{k,2}(0)$	$N_{k,2}(0)$	$\gamma_{k,3}(0)$	$N_{k,3}(0)$
				(g/cm ³)	(bcm) ⁻¹	(g/cm ³)	(bcm) ⁻¹	(g/cm ³)	(bcm) ⁻¹
1	²³⁵ ₉₂ U	235.04	4.216×10^{-2}	1.0802×10^{-4}	6.28×10^{-2}	1.6090×10^{-4}	8.91×10^{-2}	2.2828×10^{-4}	
2	²³⁸ ₉₂ U	238.05	1.305	3.3013×10^{-3}	1.952	4.9380×10^{-3}	2.758	6.9770×10^{-3}	
3	⁹² U		1.347	3.4093×10^{-3}	2.015	5.0989×10^{-3}	2.847	7.2052×10^{-3}	
4	¹⁶ ₈ O	15.999	1.3683	5.1510×10^{-2}	1.3790	5.1906×10^{-2}	1.3809	5.1978×10^{-2}	
5	¹ H	1.0079	3.97×10^{-2}	2.373×10^{-2}	3.97×10^{-2}	2.373×10^{-2}	3.97×10^{-2}	2.373×10^{-2}	
6	¹⁴ Si	28.086	0.398	8.534×10^{-3}	0.362	7.762×10^{-3}	0.312	6.690×10^{-3}	
7	²⁷ ₁₃ Al	26.982	0.270	6.026×10^{-3}	0.245	5.468×10^{-3}	0.211	4.709×10^{-3}	
8	¹² Mg	24.305	0.125	3.097×10^{-3}	0.113	2.800×10^{-3}	0.097	2.403×10^{-3}	
9	²⁶ Fe	55.847	0.286	3.084×10^{-3}	0.260	2.804×10^{-3}	0.224	2.415×10^{-3}	
10	¹⁹ K	39.098	0.0302	4.652×10^{-4}	0.0275	4.235×10^{-4}	0.0237	3.653×10^{-4}	
		$\sum_{k=3}^{10} \gamma_{k,i}$	3.509		4.081		4.780		

independent of $\gamma_{U,i}(T_0)$. The initial concentration $N_{k,i}(0)$ of nuclei, which is needed for the calculations, was calculated from the formula

$$N_{k,i}(0) = \gamma_{k,i}(0)N_A/A_k, \tag{9}$$

where $N_A = 6.022 \times 10^{23} \text{ mol}^{-1}$ is the Avogadro number and A_k is the atomic weight. For ²³⁸U and ²³⁵U we used the data of Table II; for the atoms of the rock we used the percentages by weight from Table I. The oxygen content of water was added to the oxygen of the core. The compositions of the fresh core RZ2 that we used in calculations with different initial uranium contents are shown in Table III. Although the accuracy of the densities of some elements in this table is only a few percent, the values of $N_{k,i}(0)$ are given with four decimal places for reproducibility of results.

It follows from Table II that the enrichment of isotope ²³⁵U [$\zeta_5(0) = \gamma_5(0)/\gamma_U(0)$] was $\zeta_5(0) = 3.1\%$ 1.8 billion yrs ago. Uranium of such enrichment is used in Russian WWR (water-water reactor) reactors of atomic power stations. Since the ratio of nuclei U/H is about equal and the sizes of both reactors are comparable, one can immediately and without any calculation say that a chain reaction was possible in Oklo [10].

C. Calculation of the fresh core

1. Semianalytical calculation of core RZ2

Consider first the bare reactor without reflector. Since the reactor is large compared with the neutron migration length M , one can apply the single-group diffusion theory [39,40]. For a stationary neutron flux $\Phi(\vec{r})$ the following equation holds:

$$\left[-\nabla^2 + \frac{1}{M^2} \right] \Phi(\vec{r}) = \frac{K_\infty}{K_{\text{eff}}M^2} \Phi(\vec{r}), \tag{10}$$

$$\Phi\left(\pm \frac{H}{2}\right) = \Phi(R) = 0.$$

The solution $\Phi(\vec{r})$ that satisfies this equation with boundary conditions (10) is

$$\Phi(\vec{r}) = \Phi_0 \cos\left(\frac{\pi}{H}x\right) J_0\left(\frac{2.405r}{R}\right), \tag{11}$$

where $J_0(Br)$ is the zeroth Bessel function, and the effective multiplication factor is

$$K_{\text{eff}} = \frac{K_\infty}{1 + M^2 B^2}; \quad B^2 = B_H^2 + B_R^2; \tag{12}$$

$$B_H = \frac{\pi}{H}; \quad B_R = \frac{2.405}{R}.$$

In Table IV, we show the two constants, $K_{\infty,i}$ and M_i^2 , calculated with codes MCNP4C and MCU-REA for three different cores [44,45]. These constants are needed to calculate $K_{\text{eff},i}$ by formula (12). The values of $K_{\infty,i}$ calculated for one and the same composition differ by a few tenths of a percent; the values of $M_i^2 = K_{\infty,i}\tau_i + L_i^2$ differ by a few percent. In row 9 of Table IV, we show the values of $K_{\text{eff},i}^{(1)}$ calculated with the approximate formula (12). They are smaller than the direct calculations using the Monte Carlo code (row 1). The difference in reactivity amounts to $\Delta\rho_1 = -(0.2-0.3)\%$. The diffusion length in the fresh core is $L = 1.6-2.1$ cm, and the total migration length is $M = 6-7$ cm. These lengths decrease with increasing uranium concentration.

The mean neutron flux, averaged over the reactor, is

$$\bar{\Phi} = \frac{1}{V} \int_V \Phi(\vec{r}) d\vec{r} = \Phi_0 \frac{4}{\pi} \frac{J_1(2.405)}{2.405} \tag{13}$$

[$J_1(2.405) = 0.51905$]. From formula (13) we get the following formula of the volume nonuniformity coefficient K_V , independent of R and H :

$$K_V = \Phi_0/\bar{\Phi} = \frac{\pi}{2} \frac{2.405}{2J_1(2.405)} = 3.638. \tag{14}$$

This formula is useful for checking the accuracy of the calculation of the spatial distribution $\Phi(\vec{r})$. The absolute value

TABLE IV. Two-group parameters of the fresh cores of a cylindrical bare reactor with different uranium content i . Thickness of the core $H = 1$ m; average temperature in the core $T = 300$ K; density of water $\gamma_{\text{H}_2\text{O}} = 0.355$ g/cm³ [44].

Variant of the core	1		2		3	
Relative initial density of U in the ore $Y_{U,i}(0)$, %	38.4		49.42		59.6	
Radius of the active core R (m)	9.9		8.1		6.8	
Computer code	MCNP4C	MCU-REA	MCNP4C	MCU-REA	MCNP4C	MCU-REA
K_{eff}	1.0965(1)	1.0971(1)	1.1238(2)	1.1271(1)	1.1247(2)	1.1306(1)
K_{∞}	1.1501(2)	1.1499(2)	1.1750(2)	1.1771(1)	1.1721(1)	1.1769(1)
Leakage of fast (\mathcal{L}_F) ($E > 0.625$ eV)	4.02×10^{-2}	3.96×10^{-2}	3.84×10^{-2}	3.76×10^{-2}	$3.63(1) \times 10^{-2}$	3.55×10^{-2}
Leakage of thermal (\mathcal{L}_{th}) ($E < 0.625$ eV)	0.434×10^{-2}	0.435×10^{-2}	0.317×10^{-2}	0.316×10^{-2}	$0.230(1) \times 10^{-2}$	0.230×10^{-2}
Square of diffusion length $L^2 = \mathcal{L}_{\text{th}} / [(1 - \mathcal{L}_{\text{th}}) B_{00}^2]$ (cm ²)	4.4	4.4	3.2	3.2	2.3	2.3
Age $\tau = -\log(1 - \mathcal{L}_F) / B_{00}^2$ (cm ²)	41.3	40.7	39.3	38.5	37.0	36.1
Total migration area $M^2 = K_{\infty} \tau + L^2$ (cm ²)	51.9	50.2	49.4	48.5	45.8	44.8
$B_{00}^2 = \pi^2 / H^2 + 2.4048^2 / R^2$ (cm ⁻²)	0.99292×10^{-3}		0.99584×10^{-3}		0.99954×10^{-3}	
$\pi^2 / H^2 = 0.98696 \times 10^{-3}$ (cm ⁻²)						
$K_{\text{eff}}^{(1)}$ by Eq. (12)	1.0937(2)	1.0943(2)	1.1199(2)	1.1228(1)	1.1208(2)	1.1265(1)
Account of Eq. (12) $\Delta\rho_{\text{ap}} - \rho_{\text{eff}}$, %	-0.23(2)	-0.23(2)	-0.31(2)	-0.34(2)	-0.31(3)	-0.32(1)

of the mean neutron flux for $P_p = 2.5 \times 10^{-2}$ MW is equal to

$$\bar{\Phi} = \varphi \frac{\nu_f}{E_f} P_p = 1.88 \times 10^{15} \text{ n/s } \varphi, \quad (15)$$

where φ is the neutron flux per cm² and one fast fission neutron which is calculated with the Monte Carlo code; $\nu_f / E_f = 7.5 \times 10^{16}$ n/MWs is the number of fast neutrons per MW per second (E_f is the fission energy; ν_f is the number of fast neutrons per fission). For thermal neutrons, formula (15) holds with φ_{th} . The mean thermal neutron flux with energies $E_n < 0.625$ eV is very small: in the case of $Y_{U,2}(0) = 49.4\%$, it is $\bar{\Phi}_{\text{th}} = 0.63 \times 10^8$ n/cm² s. The thermal flux in the center of the core is $\Phi_{\text{th}}^{\text{max}}(0) = 2.00 \times 10^8$ n/cm² s. The total mean flux, integrated over all energies, is equal to $\bar{\Phi} = 3.9 \times 10^8$ n/cm² s. These results were found using code MCU-REA. The results of calculations using other Monte Carlo codes are similar (see Table V). The low neutron flux determines the specifics of the function of the reactor.

As a reflector, one can assume the same core but without uranium. The analytical calculations for the reactor with reflector are more cumbersome. Therefore, we used numerical

 TABLE V. Calculations of the total flux $\bar{\Phi}_{\text{tot}}$ averaged over the bare reactor and average thermal flux $\bar{\Phi}_{\text{th}}$ ($E_n < 0.625$ eV) for the fresh core using three different codes. $Y_{02}(0) = 49.4\%$ U in the dry ore; $\omega_{\text{H}_2\text{O}}^0 = 0.355$; $P_C = 0.1$ MPa; $T = 300$ K; $P_p = 25$ kW.

	MCU-REA	MCB	MCNP4C
K_{eff}	1.1271(1)	1.1237(1)	1.1243(2)
$\bar{\Phi}_{\text{tot}}$ ($\times 10^8$ n/cm ² s)	3.9	4.4	4.4
$\bar{\Phi}_{\text{th}}$ ($E_n < 0.625$ eV) ($\times 10^8$ n/cm ² s)	0.63	0.64	0.63

methods for these calculations. The results are shown in Table VI. Both Monte Carlo programs give values of the reactivity reserve for variant 1 of the core which coincide within the statistical accuracy. The difference of reactivity is 0.26% for core RZ2 of the bare reactor and 0.46% for core 3. For the reactor with reflector, the difference is smaller: 0.20% and 0.39%, respectively. Since the migration length is small, a reflector of thickness $\Delta = 0.5$ m is practically infinite; the results for a reflector of thickness $\Delta = 0.5$ m coincide with those for $\Delta = 1$ m. Compared with the bare reactor, the reflector makes a contribution of $\delta\rho(\Delta) = 0.8\text{--}0.9\%$. This contribution drops with increasing uranium content in the core. The cold reactor with a fresh core is strongly overcritical, since temperature and void effects have not yet been taken into account, also the initial strong absorbers burn up rapidly afterwards. In Table VII, we show the neutron capture in the infinite fresh core per fast fission neutron. Capture by ^{235}U amounts to 55.7% and by ^{238}U to 33.8%. These are followed by hydrogen (3.9%), iron (3.8%), silicon (0.8%), etc. Code MCNP4C gives similar values.

2. Power effect

Reactor Oklo is controlled by the core temperature T_C [46]. During heating, the water was driven out of the core until the multiplication factor was equal to unity. At first the large overcriticality was compensated by the power effect, which is the sum of the temperature and void effects. In Table VIII and Fig. 2, we show the dependence of the water density on the temperature for several pressures. At a pressure of 100 MPa in the Oklo reactor and $T_C = 700$ K, the density of water is 65% of its value for $T_C = 300$ K and normal pressure. In this case, the difference between crystalline and free water apparently disappears. The power effect is shown in Fig. 3

TABLE VI. Calculation of K_{eff} and ρ_i for cylindrical core of thickness $H_0 = 1$ m and radius R for three different initial contents of uranium in the dry ore. Density of water $\gamma_{\text{H}_2\text{O}} = 0.355$ g/cm³; $T = 300$ K [44].

Variant of core	1			2			3		
Relative initial density of U in core $Y_{U,i}(0)$ (%)	38.4			49.4			59.6		
Specific weight of the dry ore $\gamma_i(0)$ (g/cm ³)	3.51			4.08			4.78		
Radius of active core R (m)	9.85			8.07			6.78		
Thickness of reflector Δ (m)	0	0.5	1	0	0.5	1	0	0.5	1
MCNP4C:									
K_{eff}	1.0965(1)	1.1077(1)	1.1077(1)	1.1238(2)	1.1357(2)	1.1352(1)	1.1247(2)	1.1353(2)	1.1357(1)
Reactivity $\rho_i(\%)$	8.80(1)	9.72(1)	9.72(1)	11.02(2)	11.95(2)	11.91(1)	11.09(2)	11.92(2)	11.95(1)
$\delta\rho_i(\Delta) = \rho_i(\Delta) - \rho_i(0)$	0	0.92(1)	0.92(1)	0	0.93(3)	0.89(2)	0	0.83(3)	0.86(1)
MCU-REA:									
K_{eff}	1.0971(1)	1.1080(1)	1.1080(1)	1.1271(1)	1.1383(1)	1.1381(1)	1.1306(1)	1.1407(1)	1.1405(1)
Reactivity $\rho_i(\%)$	8.85(1)	9.75(1)	9.75(1)	11.28(1)	12.15(1)	12.13(1)	11.55(1)	12.33(1)	12.32(1)
$\delta\rho_i(\Delta) = \rho_i(\Delta) - \rho_i(0)$	0	0.90(1)	0.90(1)	0	0.87(1)	0.85(1)	0	0.78(1)	0.77(1)

[44,45]. Near $T_C = 700$ K, all the $K_{\text{eff},i}$ become equal to 1. Therefore, we can assume $T_C \simeq 700$ K as the most likely temperature of the fresh active core (we neglect the small difference between the temperatures of fuel and water). For the variant of the composition of core RZ2, the power effect is $\Delta\rho_P = -11.6\%$. The void effect at 700 K accounts for 73% of this value, and the temperature effect for 27%. These results were obtained with code MCNP4C. Code MCU-REA gives similar values. In Table IV, we show the numerical values of $K_{\text{eff}}(T_C)$, $K_\infty(T_C)$, and $M^2(T_C)$, calculated with code MCU-REA for the bare reactor with core composition $i = 2$. With increased temperature, $K_\infty(T_C)$ drops and M^2 and the leakage increase (Fig. 4).

To determine the spread of results depending on the uncertainty of the initial composition of the core, the calculations were carried out over a wide range of uranium [$Y_{U,i}(0) = 39.4\text{--}59.6\%$ by weight] and of water ($\omega_{\text{H}_2\text{O}}^0 = 0.355\text{--}0.455$) contents in the ore. For reliability, the calculations for the bare reactor were carried out with two codes: MCU-REA and MCNP4C. The results were additionally controlled by the single-group formula (12) with the parameters shown in Fig. 4. The calculations of the T_C dependence of K_{eff} for variants 2 and 3 are similar (Fig. 3). For variant 1 with lower uranium content [$Y_{U1}(0) = 38.4\%$ by weight], the curve $K_{\text{eff}}(T_C)$ lies visibly lower. For a water content of $\omega_{\text{H}_2\text{O}}^0 = 0.455$, the curve for variant 2 lies significantly higher. As a result, the core temperature at which the reactor became critical was $T_C = 725$ K with a spread of ± 55 K. Taking account of the fuel

TABLE VII. Absorption $\langle \Sigma_a \Phi V \rangle_k$ and absorption with fission $\langle \Sigma_f \Phi V \rangle_k$ for infinite medium, normalized to one capture, for core variant $Y_{U,i}(0) = 49.4\%$ U; $T = 300$ K; in %.

	MCNP4C		MCU-REA	
	$\langle \Sigma_a \Phi V \rangle$	$\langle \Sigma_f \Phi V \rangle$	$\langle \Sigma_a \Phi V \rangle$	$\langle \Sigma_f \Phi V \rangle$
²³⁵ U	55.6	45.9	55.7	46.0
²³⁸ U	34.0	2.1	33.8	2.1
O	0.7		0.7	
H	4.0		3.9	
Si	0.8		0.8	
Al	0.7		0.7	
Mg	0.1		0.1	
Fe	3.7		3.8	
K	0.5		0.5	
$\sum \langle \Sigma_a \Phi V \rangle$	100.0	48.0	100.0	48.1

TABLE VIII. Temperature dependence of water density at $P_C = 100$ MPa in the core.

$T(K)$	$\gamma_{\text{H}_2\text{O}}(\text{g/cm}^3)$	$\omega_1 \gamma_{\text{H}_2\text{O}}(\text{g/cm}^3)$	$\omega_2 \gamma_{\text{H}_2\text{O}}(\text{g/cm}^3)^a$
300	1.037	0.368	0.472
350	1.014	0.360	0.461
390	0.994	0.353	0.452
400	0.982	0.348	0.447
500	0.900	0.319	0.409
536	0.864	0.307	0.393
540	0.859	0.306	0.391
600	0.792	0.281	0.360
700	0.651	0.231	0.296
800	0.482	0.171	0.219
900	0.343	0.122	0.156

^a $\omega_1 = 0.355$; $\omega_2 = 0.455$.

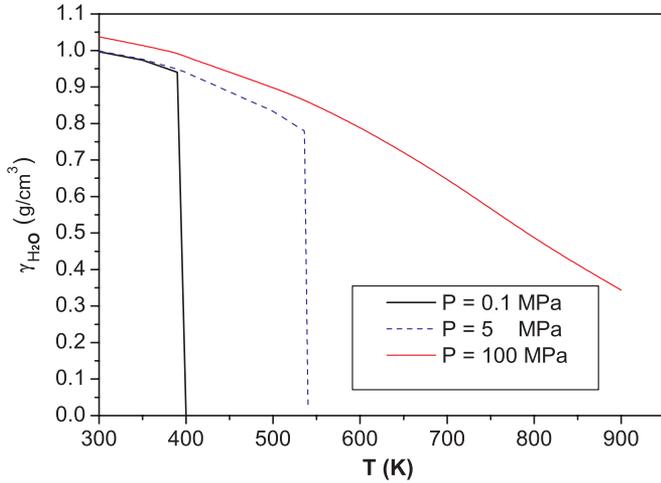


FIG. 2. (Color online) Dependence of water density γ_{H_2O} on temperature T for different pressures P .

burn-up and slogging of the reactor, a loss of reactivity takes place. This leads to a drop of T_C . The calculations of burn-up are presently continuing.

The reactor could have worked also in a pulsating mode: when the temperature exceeded 710 K, then the unbound water was boiled away, and the reactor stopped on account of the void effect. Then the water returned, and it started to work again [34,46–48]. However, a detailed analysis of the pulsating mode of operation of the reactor is outside the scope of the present paper.

3. Computational problems in calculations of large reactors

When making Monte Carlo calculations of the neutron flux in large reactors, one encounters certain difficulties. In such

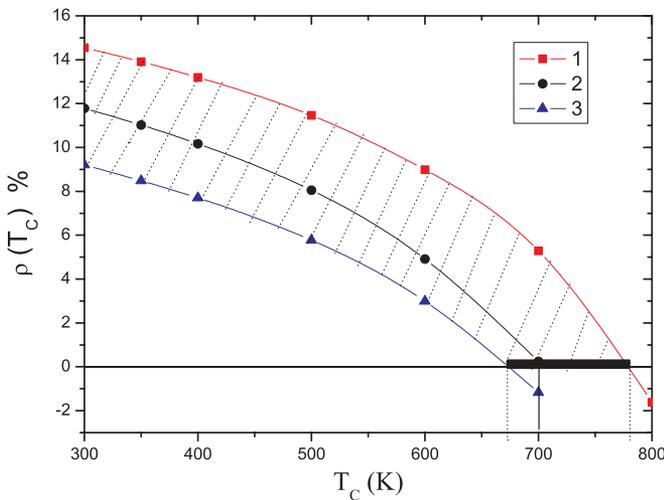


FIG. 3. (Color online) Power effect [44,45]. Dependence of reactivity ρ (in %) of the fresh core of the Oklo reactor on temperature T_C at a pressure of $P_C = 100$ MPa for three different initial compositions of the active core and different proportions of water: 1: 49.4 vol.% U in ore, $\omega_{H_2O}^0 = 0.455$; 2: 49.4 vol.% U in ore, $\omega_{H_2O}^0 = 0.405$; 3: 38.4 vol.% U in ore, $\omega_{H_2O}^0 = 0.355$. Calculations were done using code MCU-REA.

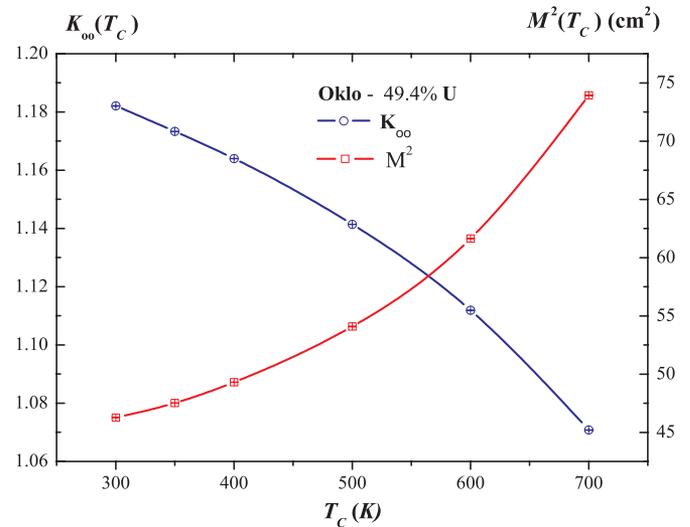


FIG. 4. (Color online) T_C dependence of $K_{\infty}(T_C)$ and $M^2(T_C)$ for a core containing 49.4 vol.% of uranium in the present-day dry ore and a water content of $\omega_{H_2O}^0 = 0.355$ at $T_C = 300$ K and $P_C = 0.1$ MPa. Calculations using code MCU-REA [44,45].

reactors, many generations are produced before a neutron that was created in the center of the reactor reaches its boundary. This time depends on the relation between the migration length and the size of the reactor [49]. At $T_C = 300$ K, these values are for core $i = 2$ equal to $M = 7$ cm and $R = 8.1$ m. Some 230 generations are needed before a centrally produced neutron reaches the boundary, detects the boundary condition, and returns to the center. To reproduce the spatial distribution of the neutron flux with sufficient accuracy, one must calculate tens of such journeys. Analyzing the solution of the time-dependent diffusion equation, one finds that over 6000 cycles are needed to get the fundamental harmonic with an accuracy of a few percent. Experience with such calculations shows that one needs $(5-10) \times 10^3$ histories per cycle in order to keep an acceptable statistical accuracy. Thus, we needed $(4-6) \times 10^7$ neutron trajectories for our calculations. For several hundred calculations, we explored an order of 10^{10} trajectories taking up several months of continuous work on a modern personal computer cluster. In spite of such a large volume of calculations, we could not find the volume nonuniformity coefficient K_V of the neutron flux with good accuracy. To do these calculations, we had to divide the core into tens of volume elements which led to a reduction of the statistical accuracy in each of them. As a result, the value of K_V in formula (14) was reproducible with an accuracy not better than 10%. This is obviously insufficient to carry out the calculation of the burn-up that depends on the magnitude of the absolute flux in different parts of the core. One must admit that the Monte Carlo method is not suitable for the calculation of large reactors, and one must resort to different approaches.

The reactor neutron spectrum below 0.625 eV is needed in order to average the cross sections of strong absorbers (e.g., $^{149}_{62}\text{Sm}$). The spectrum for three compositions of the fresh core without reflector, calculated with code MCNP4C for $T_C = 300$ K, is shown in Fig. 5. For comparison, we also show in Fig. 5 the Maxwell neutron spectrum that was used by all

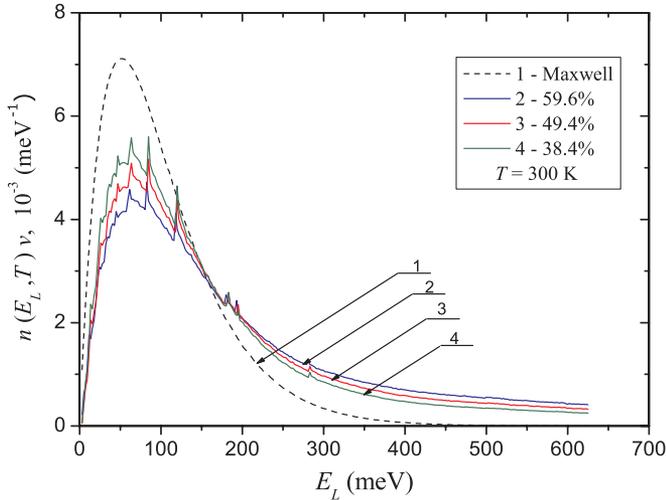


FIG. 5. (Color online) Neutron spectrum in the bare fresh core RZ2 at different initial uranium concentrations (water content $\omega_{\text{H}_2\text{O}}^0 = 0.355$). Calculations using code MCNP4C [44,45].

previous authors to average the $^{149}_{62}\text{Sm}$ cross section [7–12]. The spectra are significantly different. The Maxwell spectrum has a much higher peak but is exponentially small above 0.3 eV where the reactor spectrum is a Fermi distribution. In our calculations, we have used the Nelkin model of water which automatically takes account of the chemical bond of hydrogen nuclei. Calculations at other values of T_C yield similar results.

III. VARIATION OF FUNDAMENTAL CONSTANTS

The Oklo reactor is an instrument that is sensitive to the neutron cross sections in the distant past. By comparing them with current values, one can estimate how constant they, and hence also the fundamental constants, are in time [7–10].

A. Early approaches

In 1935, E. A. Miln [50] posed the question: How do we know that the fundamental constants are actually constant in time? He thought that the answer could be found only by experiment. A little later, D. Dirac proposed that originally all constants were of one order of magnitude, but that the gravitational constant dropped at a rate of $\dot{G}/G \sim -t_0^{-1}$ during the lifetime t_0 of the universe [51,52]. In 1967, G. Gamov suggested that, on the contrary, the electromagnetic constant is increasing: $\dot{\alpha}/\alpha \sim t_0$ [53]. Both hypotheses were wrong because they contradicted geological and paleobotanical data from the early history of the Earth. Without entering into a detailed discussion of these and many other later publications on this subject, it seems that there is a problem with the experimental limit on the rate of change of the fundamental constants (see the early review by F. Dyson [54]).

The authors of Refs. [7–10] noticed that the sensitivity to variations of the nuclear potential increases by several orders of magnitude if one considers neutron capture. Owing to the

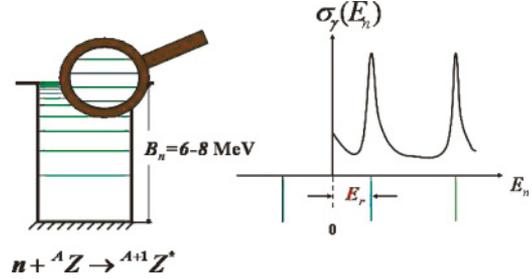


FIG. 6. (Color online) Strong absorber as a sensitive detector of a variation of E_r [55]. Left: energy level density of the compound nucleus $n + {}^A Z \rightarrow {}^{A+1} Z^*$. Right: resonances in the cross section of the $n\gamma$ reaction. The capture cross section behaves like $\sigma_\gamma \simeq (\Gamma_\gamma/E_r)^2$, where E_r is the distance from the resonance and Γ_γ is its width. Neutron capture is strongly affected by a shift of ΔE_r .

sharp resonances of the absorption cross section, the nucleus is a finely tuned neutron receiver. A resonance shifts on the energy scale with changes of the nuclear potential similarly as the frequency of an ordinary radio receiver shifts when the parameters of the resonance circuit are changed (Fig. 6) [55]. Qualitatively, one can understand the absence of a significant shift of the near-threshold resonances on the grounds that all strong absorbers are highly burned up in the Oklo reactor, and weak absorbers are burned up weakly (Fig. 7) [10,56]. Holes in the distributions are seen for strong absorbers: $^{149}_{62}\text{Sm}$, $^{151}_{63}\text{Eu}$, $^{155}_{64}\text{Gd}$, $^{157}_{64}\text{Gd}$. The depth of burn-up, calculated using the present absorption values, are in satisfactory agreement with experiment, particularly if one remembers that the neutron spectrum over which one must average the cross section is not very well known. Thus, in the 1.8 billion yr since the work of the Oklo reactor, the resonances (or, in other words, the levels of the compound nuclei) have shifted by less than $\Delta E_r \simeq \Gamma_\gamma/2$ ($\Gamma_\gamma = 0.1$ eV). Therefore, the average rate of the shift did not exceed 3×10^{-11} eV/yr. This value is at least three orders of magnitude less than the experimental limit on the rate of change of the transition energy in the decay of ^{187}Re [54].

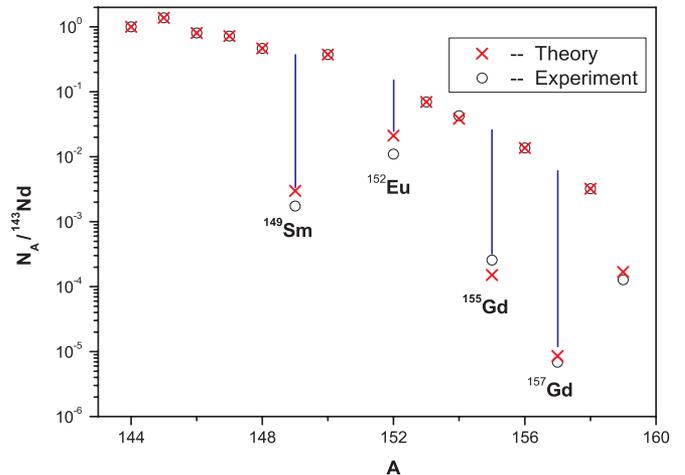


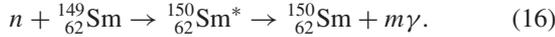
FIG. 7. (Color online) Comparison of the calculated (crosses) and measured (circles) concentrations of fission fragments N_A relative to the ^{143}Nd content for one Oklo sample [10,56].

As pointed out previously [10], there are no reliable theoretical calculations to date relating a shift of nuclear resonances to a change of fundamental constants. Such calculations would be very desirable but they cannot be done without additional assumptions about the mechanism of a change of constants (see the reviews [57]). Nevertheless it is possible to exclude the hypotheses of a power or logarithmic dependence of the constants on the lifetime of the universe, using preliminary estimates based on the Oklo data.

B. Basic formulas

1. Averaging the Breit-Wigner formula

When a slow neutron is captured by a nucleus of isotope $^{149}_{62}\text{Sm}$, then a nuclear reaction takes place with the formation of an excited intermediate compound nucleus and subsequent emission of $m\gamma$ quanta:



Near a strong S resonance, one can neglect the effect of the other resonances and describe the cross section with the Breit-Wigner formula

$$\sigma_{\gamma,\text{Sm}}(E_C) = g_0 \frac{\pi \hbar^2}{2m_n E_C} \frac{\Gamma_n(E_C)\Gamma_\gamma}{(E_C - E_r)^2 + \Gamma_{\text{tot}}^2/4}, \quad (17)$$

where $g_0 = (2J + 1)/((2S + 1)(2I + 1))$ is the statistical factor, $S = 1/2$ is the electron spin, I is the nuclear spin, and J is the spin of the compound nucleus. The full width is $\Gamma_{\text{tot}} = \Gamma_n(E) + \Gamma_\gamma$, where $\Gamma_n(E)$ and Γ_γ are the neutron and γ width, respectively. The neutron width is given by [58]

$$\Gamma_n(E_C) = \Gamma_n^0 \sqrt{\frac{E_C}{E_0}}; \quad E_0 = 1\text{eV}. \quad (18)$$

The parameters of the lowest resonances of a number of absorbers is given in Table IX.

In formula (17) the neutron energy is given in the c.m. frame: $E_C = \frac{1}{2}m_n|\vec{V}_L - \vec{V}_k|^2$. It depends on the velocities of the nucleus \vec{V}_k and the neutron \vec{V}_L in the laboratory frame and on the reduced mass m_n . The reaction rate $N_k\sigma_{\gamma,k}(E_C)V_C$ with cross section (17) and for an absorber of nuclear density N_k must be averaged over the nuclear spectra $f_k(E_k)$ and the neutron spectrum $n(E_L)$ (all spectra are normalized to 1). The

TABLE IX. Lowest resonance parameters of strong absorbers [58].

	$^{149}_{62}\text{Sm}$	$^{155}_{64}\text{Gd}$	$^{157}_{64}\text{Gd}$
Resonance energy E_r , (meV)	97.3	26.8	31.4
Neutron width at 1 eV Γ_n^0 (meV)	1.71(3)	0.635(10)	2.66(5)
Gamma width Γ_γ (meV)	60.5(6)	108(1)	106(1)
g factor	9/16	5/8	5/8
Doppler width Δ_D (meV) at $T = 700$ K	12.5	6.4	6.9

inverse nuclear burn-up time in an arbitrary point of the core is given by

$$\lambda_{\gamma,k}(T) = N_k \int d\vec{p}_k d\vec{p}_L f_k(E_k)n(E_L)\sigma_{\gamma,k}(E_C)V_C. \quad (19)$$

At high temperatures, the gas approximation is valid for heavy nuclei of the absorber. Changing to integration over the c.m. energy E_C and the neutron energy E_L and assuming a Maxwell nuclear spectrum, we get

$$\lambda_{\gamma,k}(T) = N_k \int n(E_L)\sigma_{\gamma,k}(E_C)V_C F(E_C \rightarrow E_L) dE_L dE_C, \quad (20)$$

where $F(E_C \rightarrow E_L)$ is the transformation function from the c.m. to the laboratory system (for details see Ref. [59]), that is,

$$F(E_C \rightarrow E_L) = \frac{(A+1)}{2\sqrt{\pi A T E_L}} \times \left\{ \exp \left[-\frac{A}{T} \left(\sqrt{\left(1 + \frac{1}{A}\right) E_C} - \sqrt{E_L} \right)^2 \right] - \exp \left[-\frac{A}{T} \left(\sqrt{\left(1 + \frac{1}{A}\right) E_C} + \sqrt{E_L} \right)^2 \right] \right\}, \quad (21)$$

and $A = M_A/m_n$ is the mass of nucleus A in units of the neutron mass.

Close to a resonance, we can neglect the second term in Eq. (21) and evaluate the first term in integral (20) by the saddle-point method. As a result, in the vicinity of a resonance, the integral (20) becomes

$$\lambda_{\gamma,k}(T) = N_k \frac{\pi}{2} \left(1 + \frac{1}{A}\right) \int dE_C \sigma_{\gamma,k}(E_C) V_C \int dE_L n(E_L) \Gamma \times \left[E_L - \left(1 + \frac{1}{A}\right) E_C \right], \quad (22)$$

where the Gaussian

$$\Gamma \left[E_L - \left(1 + \frac{1}{A}\right) E_C \right] = \frac{1}{\sqrt{\pi} \Delta_D} \exp \left\{ -\frac{\left[E_L - \left(1 + \frac{1}{A}\right) E_C \right]^2}{\Delta_D^2} \right\} \quad (23)$$

is normalized to unity and the Doppler width is equal to

$$\Delta_D = \left[\frac{4E_L T}{A} \right]^{1/2} = \left[\frac{4E_C T}{A+1} \right]^{1/2}. \quad (24)$$

The values of the Doppler widths for $T = 700$ K are shown in Table IX. Since all $\Delta_D \ll \Gamma_\gamma$, function (23) can be replaced by $\delta(|E_L - A E_C/(A+1)|)$ and integral (22) becomes

$$\lambda_{\gamma,k} = N_k \frac{\pi}{2} \left(1 + \frac{1}{A}\right)^2 \int \sigma_{\gamma,k}(E_C) V_C n \left[\left(1 + \frac{1}{A}\right) E_C \right] dE_C. \quad (25)$$

The correction $2/A$ is of magnitude 1%. If the neutron spectrum is Maxwellian in the c.m. frame, then it is also Maxwellian (with reduced neutron mass) when the nuclear motion is taken into account. It can be shown that Eq. (25) is valid at all energies if the distribution of nuclei and neutrons is Maxwellian [60].

Therefore, it is not surprising that the authors of Ref. [12] did not notice any deviations from formula (25) in their numerical calculation, which took account of the thermal motion of the target nuclei (Doppler effect). However, the situation is different if the neutron spectrum is not Maxwellian. In this case, one must use formula (22) instead of the simple formula (25).

To average the capture cross section of samarium, one normalizes the cross section, integrated over the neutron flux spectrum $n(E, T)v$, traditionally not by the integrated flux but by the product of the velocity $v_n^0 = 2200$ m/s and the integrated neutron density $n(E)$ [11,12,60]:

$$\hat{\sigma}_{\gamma,k}(T) = \frac{\int \sigma_{\gamma,k}(E_L)n(E_L)v_L dE_L}{v_n^0 \int n(E_L)dE_L}. \quad (26)$$

If the cross section $\sigma_{\gamma,k}(E_L)$ has a $1/v_L$ behavior, then the integral of $\hat{\sigma}_{\gamma,k}(T)$ is constant. From formula (26) one has

$$\hat{\sigma}_{\gamma,k}(T) = \sqrt{\frac{4T}{\pi T_0}} \sigma_{\gamma,k}(T), \quad (27)$$

where $T_0 = 300$ K = 25.9 meV. Useful also is the relation [12]

$$\sigma \Phi = \hat{\sigma} \hat{\Phi}, \quad \text{where} \quad \hat{\Phi} = \sqrt{\frac{\pi T_0}{2 T}} \Phi. \quad (28)$$

We have evaluated the cross section $\hat{\sigma}_{\gamma,\text{Sm}}(T)$ of ^{149}Sm without recourse to any approximations. For $\lambda_{\gamma,\text{Sm}}(T)/(N_{\text{Sm}}v_n^0)$ we have

$$\hat{\sigma}_{\gamma,\text{Sm}}(T) = \frac{\sqrt{\pi} \int dE_k dE_L f_{\text{Sm}}(E_k) \sigma_{\gamma,\text{Sm}}(E_C) V_C n(E_L)}{v_n^0 \int dE_L n(E_L)}. \quad (29)$$

For the calculations, we used the computer package MATHEMATICA [61]. In Fig. 8 we show the values of $\hat{\sigma}_{\gamma,\text{Sm}}(T, \Delta E_r)$ at six temperatures $T = 300$ – 1000 K for a shift of the resonance position $\Delta E_r = \pm 0.2$ eV. The curves have a maximum at negative shifts of the resonance; the maximum of the curves is higher at lower temperature T . At the point $\Delta E_r = 0$ and at $T = 293$ K, the cross section calculated as the contribution of the closest resonance is $\sigma_{\gamma,\text{Sm}}(293 \text{ K}) = 39.2$ kb. The contribution of higher positive resonances is $\sigma_{\gamma,\text{Sm}}^+(293 \text{ K}) = 0.6$ kb, and that of negative ones is $\sigma_{\gamma,\text{Sm}}^-(293 \text{ K}) = 0.3$ kb [58]. The total cross section (as measured on a neutron beam) is $\sigma_{\gamma,\text{Sm}}^{\text{tot}}(293 \text{ K}) = 40.1$ kb. At small energy shifts ΔE_r , $\sigma_{\gamma,\text{Sm}}^+$ and $\sigma_{\gamma,\text{Sm}}^-$ practically do not change. In Refs. [11,12], the total cross section of 40.1 kb has been used instead of the single resonance one, 39.2 kb. Therefore, the curves in Refs. [11,12] are higher by 40.1 kb/39.2 kb, i.e., by 2.5%.

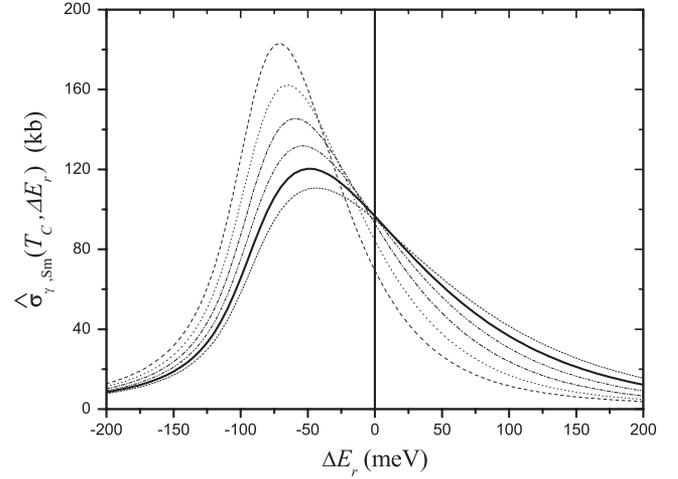


FIG. 8. Dependence of the cross section $\hat{\sigma}_{\gamma,\text{Sm}}$, averaged over a Maxwell neutron spectrum, on the resonance shift ΔE_r and on the temperature $T = 300$ – 800 K, Eq. (29). The curves are for fixed temperatures with intervals of 100 K. The highest curve is for $T = 300$ K.

2. Taking account of the reactor spectrum

In Fig. 9 we show the results of calculating $\hat{\sigma}_{\gamma,\text{Sm}}(T_C, \Delta E_r)$ with the Maxwell spectrum replaced by the reactor spectrum $n_R(E_L, T_C)$. The central curve 2 is the result of the calculation using code MCNP4C for the fresh core with $Y_{\text{U}2}(0) = 49.4\%$ U in the dry ore and with $\omega_{\text{H}2\text{O}}^0 = 0.405$ at $T = 725$ K. For comparison, we also show the cross section averaged over the

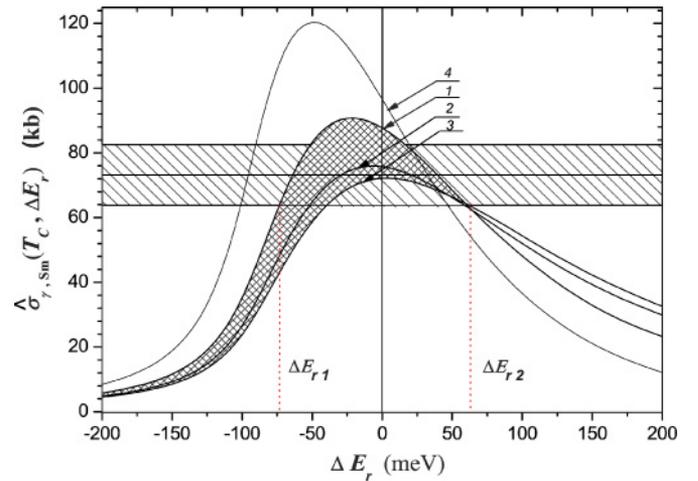


FIG. 9. (Color online) Dependence of thermal neutron capture cross section of ^{149}Sm on core temperature T_C and on resonance shift ΔE_r : $\hat{\sigma}_{\gamma,\text{Sm}}(T, \Delta E_r)$ [44,45]. The curves are for cross sections, averaged over the reactor spectrum of the fresh core at three different initial states: 1: $Y_{\text{U}2}(0) = 38.4$ vol.% U, $\omega_{\text{H}2\text{O}} = 0.355$, $T_C = 670$ K; 2: $Y_{\text{U}2}(0) = 49.4$ vol.% U, $\omega_{\text{H}2\text{O}} = 0.405$, $T_C = 725$ K; 3: $Y_{\text{U}2}(0) = 49.4$ vol.% U, $\omega_{\text{H}2\text{O}} = 0.455$, $T_C = 780$ K, $P_C = 100$ MPa. For comparison we show the cross section averaged over the Maxwell spectrum (curve 4) for initial composition $Y_{\text{U}2}(0) = 38.4$ vol.% U, $\omega_{\text{H}2\text{O}} = 0.405$, $T_C = 725$ K. Shown is the error corridor of measured values $\hat{\sigma}_{\gamma,\text{Sm}}^{\text{Exp}}$ [11].

Maxwell spectrum at $T = 725$ K for the same composition of the core (curve 4). Curves 4 and 2 are significantly different, especially at negative ΔE_r : curve 2 lies distinctly lower. The maximum of curve 2 is 1.5 times lower than the maximum of curve 4. At lower temperatures, this difference is even greater. Thus we conclude that we have proved a significant *effect of the reactor spectrum on the cross section of ^{149}Sm* .

In order to determine the dependence of $\hat{\sigma}_{\gamma,\text{Sm}}(T_C, \Delta E_r)$ on the uncertainty in the initial active core composition, we calculated the values for the two outermost curves of Fig. 9. Curve 3 of this figure corresponds to an initial content of $Y_{\text{U1}}(0) = 38.4\%$ U in the ore, $\omega_{\text{H}_2\text{O}}^0 = 0.355$, and $T_C = 670$ K; curve 1 corresponds to an initial content of 49.4%, $\omega_{\text{H}_2\text{O}}^0 = 0.455$ at $T = 780$ K. Since the numerical constants are known only for values of T_C which are multiples of 100, we performed the calculations for $T_C = 600, 700, 800$ K and interpolated to intermediate temperatures. The broadening of curve 2 on account of the scatter of temperatures is small. Experimental data of $\hat{\sigma}_{\gamma,\text{Sm}}^{\text{Exp}}(T)$ for core 2 are presented in Ref. [11] (Table X) (see also Ref. [63,64]). The labeling of sample SC36–1418 indicates that the sample was taken from bore-hole SC36 at a depth of 14 m 18 cm. The mean value $\bar{\sigma}_{\gamma,\text{Sm}}^{\text{Exp}} = (73.2 \pm 9.4)$ kb is shown in Fig. 9. Curve 1 ($T_C = 670$ K) crosses the lower limit of $\bar{\sigma}_{\gamma,\text{Sm}}^{\text{Exp}} = 64$ kb to the left of point $\Delta E_r^{(1)} = -73$ meV, and curve 3 ($T_C = 780$ K) to the right at $\Delta E_r^{(2)} = +62$ meV. The possible shift of the resonance is therefore given by the limits

$$-73 \text{ meV} \leq \Delta E_r \leq 62 \text{ meV}. \quad (30)$$

It is important that for core RZ2 a complete set of the detailed information is available. This set should include the dependence of the uranium content, ^{235}U burn-up, etc., on the uranium density γ_{U} in the dry ore (see Fig. 1). The data should belong to the same bore-hole. For the cylindrical core, all points from the bore-hole are located at the same distance from the center of the reactor with the same neutron flux and

TABLE X. Experimental values of $\hat{\sigma}_{\text{Sm}}$ for 15 samples from Oklo core RZ2 [11,38].

		$\hat{\sigma}_{\text{Sm}}$ (kb)	
1	KN50-3548	93	[56]
2	SC36-1440	73	[62]
3	SC36-1410/3	73	[62]
4	SC36-1413/3	83	[62]
5	SC36-1418	64	[62]
6	SC39-1383	66	[27,62]
7	SC39-1385	69	[27,62]
8	SC39-1389	64	[27,62]
9	SC39-1390	82	[27,62]
10	SC39-1391	82	[27,62]
12	SC35bis-2126	57	[27,62]
13	SC35bis-2130	81	[27,62]
14	SC35bis-2134	71	[27,62]
15	SC52-1472	72	[32]
$\bar{\sigma}_{\text{Sm}} \pm \Delta \hat{\sigma}_{\text{Sm}}$		(73.2 ± 9.4)	

^{235}U burn-up. The number of points should be large enough to obtain reliable average values of the concentrations at the given place near the center of the reactor. This is the case for core RZ2 where 15 points are known. Averaging data from different bore-holes is not allowed, as the burn-up of ^{235}U and its concentration depends on the distance to the core center. The average concentrations serve as input for reactor calculations in order to describe the neutronics of the core.

This is why we chose core RZ2 for the first time calculations of the Oklo reactor spectrum.

3. Connection between ΔE_r and $\delta\alpha/\alpha$

The shift ΔE_r must be related to a variation of the fundamental constants, for instance, a shift of the electromagnetic (e.m.) constant $\alpha = 1/137.036$. This was done by Damour and Dyson, assuming that α was the only fundamental constant that could change with time [11]. The main contribution to the resonance energy E_r due to the electromagnetic interaction is given by the difference of the expectation values of the Coulomb energy of the nuclei:

$$\alpha \frac{dE_r}{d\alpha} = \alpha \frac{d}{d\alpha} (\langle H_c \rangle_1 - \langle H_c \rangle_2), \quad (31)$$

where H_c is the Coulomb part of the Hamiltonian and subscript 1 (2) refers to a highly excited state of the compound nucleus $^{150}_{62}\text{Sm}$ (ground state of $^{149}_{62}\text{Sm}$). This energy difference is related to the change of the proton density in the transition from one nucleus to the other, and it can be expressed in terms of the mean square radius of the nucleus $\langle r^2 \rangle$

$$\langle H_c \rangle_1 - \langle H_c \rangle_2 \approx -\frac{Z\alpha}{2R^3} (\langle r^2 \rangle_2 - \langle r^2 \rangle_1). \quad (32)$$

The difference between mean square radii can be extracted from the isotope effect, i.e., the change of energy of the s electrons in the transition from one nucleus to the other. As the value of the isotope effect of highly excited states of $^{149}_{62}\text{Sm}$ is not known, Damour and Dyson estimated it by isotope effect of the ground states. Using data on the isotope effect of Sm atoms (see also [65]), they found

$$\langle r^2 \rangle_2 - \langle r^2 \rangle_1 = 0.211 \pm 0.017 \text{ fm}^2. \quad (33)$$

For radius $R \approx 8.11$ fm of the $^{149}_{62}\text{Sm}$ nucleus, we get from formula (32)

$$\mathcal{M} \equiv \alpha \frac{dE}{d\alpha} = -(1.1 \pm 0.1) \text{ MeV}. \quad (34)$$

The ratio of \mathcal{M} to the resonance energy $E_r \approx 0.1$ eV is of order of 10^7 . This explains the high sensitivity of the Oklo data to changes of the fine structure constant.

Combining this value with the shift of the resonance ΔE_r in formula (30), we get

$$-5.6 \times 10^{-8} < \delta\bar{\alpha}/\alpha < 6.6 \times 10^{-8}. \quad (35)$$

Because of the negative value of \mathcal{M} , the limits on $\delta\bar{\alpha}/\alpha$ change their places. For the past time ($-T_0$), the product ($-T_0\mathcal{M}$) is positive, and hence the limits on $\delta\bar{\alpha}/\alpha \equiv \Delta E_r/(-T_0\mathcal{M})$ are restored to their previous places. Note that traditionally $\delta\bar{\alpha}/\alpha$ is defined by $\delta\alpha = (\alpha_{\text{Oklo}} - \alpha_{\text{now}})/\alpha$. This shift of α lies in

a narrower range than that in Ref. [11]. Assuming a linear change of the e.m. constant during the time T_0 , we get the following limit on the relative rate of change:

$$-3.7 \times 10^{-17} \text{ yr}^{-1} < \dot{\delta\alpha}/\alpha < 3.1 \times 10^{-17} \text{ yr}^{-1}. \quad (36)$$

Thus, within the limits given by Eq. (36), the e.m. constant changes for the fresh reactor Oklo core with zero speed, i.e., it remains constant.

C. Review of previous work

To convince ourselves of the absence of change of the ^{149}Sm cross section, we shall consider in more detail the previous work (see also the review by J. Uzan [57]).

The work of Shlyakhter (1976, 1983) [7–9]. The authors of Refs. [7–10] were the first to point out the possibility of using the data of the natural nuclear reactor Oklo to find the most precise limits on the rate of change of the fundamental constants. The most convenient data are those of the strong absorbers, e.g., ^{149}Sm . For this isotope, Shlyakhter calculated at $T = 300 \text{ K}$ the dependence of the change of the cross section on the resonance by an amount of ΔE_r : $\sigma_{\gamma, \text{Sm}}(T_C, \Delta E_r)$ (Fig. 10) [8].

He compared this curve with the experimental data available at the time: $\bar{\sigma}_{\gamma, \text{Sm}}^{\text{Exp}} = (55 \pm 8) \text{ kb}$. A possible shift of the first resonance within two standard deviations (95% confidence level) was found to be

$$\delta E_{r2}^{\text{Exp}} \leq 20 \text{ meV}. \quad (37)$$

[In going from $\bar{\sigma}_{\gamma, \text{Sm}}(T, \Delta E_r)$ to $\hat{\sigma}_{\gamma, \text{Sm}}(T, \Delta E_r)$, all values must be multiplied by 1.18 according to Eq. (27) and $\bar{\sigma}_{\gamma, \text{Sm}}^{\text{Exp}} = (65 \pm 9.5) \text{ kb}$, but this does not affect the value of $\delta E_{r2}^{\text{Exp}}$]. To estimate \mathcal{M} , Shlyakhter used data on the compressibility of the nucleus; he found $\mathcal{M} = -2 \text{ MeV}$ [8]. With a linear dependence of the change of α with time for $T_0 = 2 \times 10^9 \text{ yr}$ the limit on the rate of change of α is

$$\dot{\delta\alpha}/\alpha \leq 0.5 \times 10^{-17} \text{ yr}^{-1}. \quad (38)$$

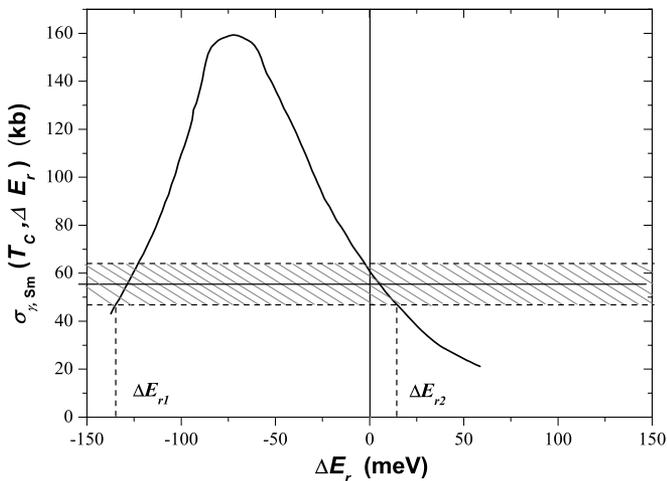


FIG. 10. Change of the thermal ($T = 25 \text{ meV} = 273 \text{ K}$) capture cross section of ^{149}Sm for a uniform shift of all resonances by ΔE_r [8].

Using the present, more accurate value of $\mathcal{M} \simeq -1.1 \text{ MeV}$, we get

$$\dot{\delta\alpha}/\alpha \leq 1 \times 10^{-17} \text{ yr}^{-1}. \quad (39)$$

It should be emphasized again that this limit was found for only one temperature: $T = 300 \text{ K}$.

The work of Petrov (1977) [10]. In this paper, the resonance shift δE_r was estimated from the shifts of the widths of a few strong absorbers. The resonances of strong absorbers lie close to a zero energy of the neutron, and the resonance energy is of the capture width: $\delta E_r \sim \Gamma_\gamma \simeq 0.1$. The capture cross section of these absorbers for thermal neutrons changes sharply when the resonance is shifted by an amount of the order of $\Gamma_\gamma/2$. The analysis of experimental data for ^{149}Sm and ^{151}Eu , taking account of a threefold standard deviation and the uncertainty of the core temperature, shows that the shift δE_r of the resonance since the activity of the Oklo reactor does not exceed $\pm 0.05 \text{ eV}$ [10]. The results of the measurement of the concentration of rare earth elements with respect to ^{143}Nd (the second branch of the mass distribution of the fission fragments) in one of the Oklo samples are shown in Fig. 7. A more conservative estimate in Ref. [10] is $|\delta E_r| \leq 50 \text{ meV}$, i.e., 2.5 times higher than Shlyakhter's estimate. Using the modern value $\mathcal{M} \simeq -1.1 \text{ MeV}$, we find for $\dot{\delta\alpha}/\alpha \leq \delta E_r/|\mathcal{M}|$

$$\dot{\delta\alpha}/\alpha = \delta E_r/|\mathcal{M}| \leq 4.5 \times 10^{-8}. \quad (40)$$

This is almost five times greater than Shlyakhter's optimistic estimate. For the rate of change $\dot{\delta\alpha}/\alpha$ we get

$$\dot{\delta\alpha}/\alpha \leq 2.5 \times 10^{-17} \text{ yr}^{-1}. \quad (41)$$

This is less by a factor of 2 than the limit found 20 years later by Damour and Dyson [11]. The reason for this discrepancy is the use of only one temperature ($T_C = 300 \text{ K}$). Although the dependence of ΔE_r on T_C was noted in Ref. [10], no calculations of the effect of the temperature were carried out.

The work of Damour and Dyson (1996) [11]. The dependence $\Delta E_r(T_C)$ was analyzed 20 years later in Ref. [11]. They repeated the analysis of Shlyakhter and came to the conclusion that it was correct. They also updated Shlyakhter's data in three directions:

- (i) They employed a large amount of experimental data (see Table X).
- (ii) They accounted for the great uncertainty of the reactor temperature, $T_C = 450\text{--}1000 \text{ }^\circ\text{C}$ (Fig. 11). As a result, they made a conservative estimate of the mean shift of the resonance

$$-120 \leq \bar{\Delta E}_r \leq 90 \text{ meV}. \quad (42)$$

The range of the shift $\Delta E_{r1} - \Delta E_{r2} = 210 \text{ meV}$ is 1.5 times greater than the range of the shift in our paper.

- (iii) They calculated the value $\mathcal{M} = -(1.1 \pm 0.1) \text{ MeV}$, but used $\mathcal{M} = -1 \text{ MeV}$. For $\dot{\delta\alpha}/\alpha$, they found

$$-9.0 \times 10^{-8} \leq \dot{\delta\alpha}/\alpha \leq 12 \times 10^{-8}. \quad (43)$$

This leads to the following limits on the rate of change $\dot{\delta\alpha}/\alpha$:

$$-6.7 \times 10^{-17} \leq \dot{\delta\alpha}/\alpha \leq 5.0 \times 10^{-17} \text{ yr}^{-1}. \quad (44)$$

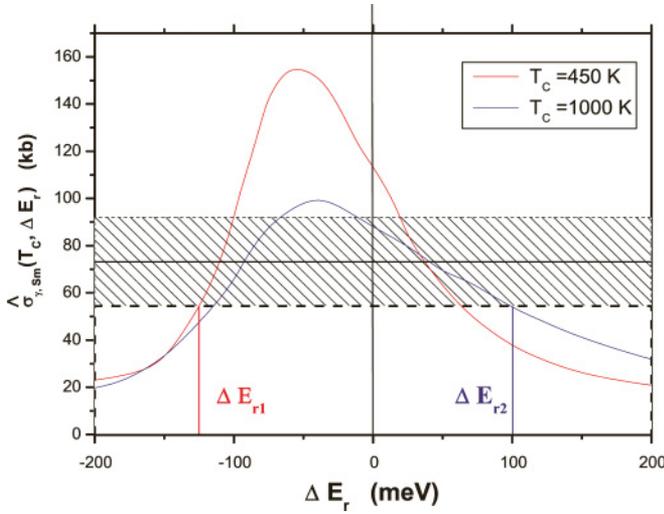


FIG. 11. (Color online) Dependence of the thermal neutron capture cross section on the resonance shift ΔE_r for the two temperatures of the Maxwell neutron spectrum (Damour and Dyson [11]). Energies of the crossing of the lower limit of data are $\Delta E_{r1}^M = -120$ meV and $\Delta E_{r2}^M = 90$ meV.

Since ^{149}Sm burns up 100 times faster than ^{235}U , the only ^{149}Sm found in the stopped reactor is that which was produced immediately before the end of the cycle. As a consequence, Damour and Dyson emphasized that one must know the detailed distribution of the nuclear reaction products of the end of the cycle to make a detailed analysis.

The work of Fujii *et al.* (2000, 2002) [12,64]. Cores other than RZ2 were also intensively investigated (see Refs. [66,67]). From core RZ9, five samples were taken with high concentrations of uranium and high burn-up of ^{235}U in the one bore-hole. The concentration of rare earth elements in these samples (^{149}Sm included) was also measured. For core RZ10 (located 150 m deeper than RZ2), four additional points are known, and one more point for core RZ13. Finally, three more samples which obey required conditions were obtained from the Bangoombe reactor, which is 8 km away from Oklo reactors. Unfortunately, authors of Refs. [66,67] did not extract neutron absorption cross sections.

The experimental data on the measurement of $\hat{\sigma}_{\gamma,k}$ of strong absorbers were presented in the papers of Fujii *et al.* Of five experimental points, four are from core RZ10 (Table XI) and

TABLE XI. Experimental values of $\hat{\sigma}_{\gamma,k}^{\text{Exp}}$ (kb) for strong absorbers, measured in the middle of core RZ10, located at a depth of 150 m [12].

	^{149}Sm	^{155}Gd	^{157}Gd
SF84-1469	85.6	30.9	83.3
SF84-1480	96.5	16.8	8.0
SF84-1485	83.8	17.8	14.3
SF84-1492	99.0	36.7	73.7
$\bar{\sigma}_k$	90.7	25.6	44.8
$\pm \Delta \sigma_k$	8.2	9.8	39.2

only one from another core, RZ13, which we have omitted. As we do not have any detailed data on the size and composition of core RZ10, we shall assume them to be similar to those of core RZ2 with the uranium concentration corresponding to that measured in core RZ10. For ^{149}Sm the mean value of the four points of Table XI is

$$\bar{\sigma}_{\gamma,Sm}^{\text{Exp}} = (90.7 \pm 8.2) \text{ kb}. \quad (45)$$

This value is noticeably greater than for core RZ2 [(72.3±9.4) kb (see Sec. III B2)]. The error bars of both values do not even touch, and their difference remains significant. For increased reliability, the number of measurements for core RZ10 should be increased. Possibly this core finished its cycle at a lower temperature.

In Fig. 12, we show the dependence of $\hat{\sigma}_{\gamma,Sm}(T_c, \Delta E_r)$ on the shift of the resonance for a Maxwell distribution [12,64]. The authors estimated (on the grounds of indirect considerations) the uncertainty in $T_c = 180\text{--}400^\circ\text{C} = 453\text{--}673$ K. They also show the experimental data of formula (45). The intersection of the limiting curves with the lower limit $\hat{\sigma}_{\gamma,Sm}^{\text{Exp}} = 82.5$ kb yields the following possible shift of ΔE_r :

$$-105 < \Delta E_r < +20 \text{ meV}. \quad (46)$$

In Fig. 12, we also show for comparison the curves $\hat{\sigma}_{\gamma,Sm}(T_c, \Delta E_r)$ for the reactor spectrum of the fresh core at $T_c = 400^\circ\text{C}$, $Y_{U1}(0) = 38.4\%$ U in the ore, $\omega_{\text{H}_2\text{O}}^0 = 0.355$, and $P = 100$ MPa. The possible shift of ΔE_r for the experimental data of formula (45) lie in a narrower interval than those in [11]:

$$-120 \leq \Delta E_r \leq 20 \text{ meV}. \quad (47)$$

From Eq. (46) and using $\mathcal{M} = -1.1$ MeV we get

$$-1.8 \times 10^{-8} \leq \bar{\delta\alpha}/\alpha \leq 9.5 \times 10^{-8} \quad (48)$$

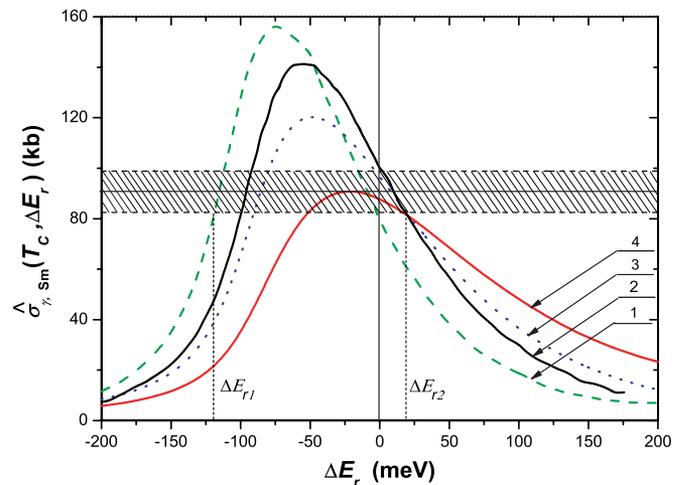


FIG. 12. (Color online) Dependence $\hat{\sigma}_{\gamma,Sm}(T_c, \Delta E_r)$ for the Maxwell distribution at $T_c = 450, 570, 670$ K (curves 1–3) (Fujii *et al.* [12,64]). Data correspond to Table XI; $\hat{\sigma}_{\gamma,Sm}^{\text{Exp}} = 90.7 \pm 8.2$ kb, $\Delta E_{r2}^M = 20$ meV. For comparison, we show curve 4 for the reactor spectrum at $T_c = 670$ K; $Y_{U1}(0) = 38.4$ vol.% U; $\omega_{\text{H}_2\text{O}}^0 = 0.355$; $P_c = 100$ MPa.

TABLE XII. Limits on the rate of α variation, based on the data of ^{149}Sm contents in the Oklo reactor; $z = 1.8 \times 10^9 \text{ yr}/13.7(2) \times 10^9 \text{ yr} = 0.131(2)$; $t_0 = 13.7(2)$ [75]; and data obtained by other methods.

Lab	Authors, year	$\Delta E_r, \overline{\delta\alpha}/\alpha, \dot{\delta\alpha}/\alpha$	Comments core, spectrum, T_C
LNPI, Gatchina Russia	A. Shlyakhter, 1976 [8]	$\Delta E_r \leq 20 \text{ meV}$ $\dot{\delta\alpha}/\alpha \leq 0.5 \times 10^{-17} \text{ yr}^{-1}$	RZ2, Maxwell, 300 K
LNPI,Gatchina Russia	Yu. Petrov, 1977 [10]	$\Delta E_r \leq 50 \text{ meV}$ $\dot{\delta\alpha}/\alpha \leq 2.5 \times 10^{-17} \text{ yr}^{-1}$	RZ2, Maxwell, 300 K
Princeton, USA	T. Damour and F. Dyson, 1996 [11]	$\Delta E_r \leq 90 \text{ meV}$ $\dot{\delta\alpha}/\alpha \leq 5.0 \times 10^{-17} \text{ yr}^{-1}$	RZ2, Maxwell, 450–1000 K
Univ.Tokyo, Tokyo,Japan	Ya. Fujii <i>et al.</i> 2000 [12]	$\Delta E_r \leq 20 \text{ meV}$ $\dot{\delta\alpha}/\alpha \leq 1.0 \times 10^{-17} \text{ yr}^{-1}$	RZ10, Maxwell,470–670 K
PNPI,Gatchina Russia	This paper	$\Delta E_r \leq 62 \text{ meV}$ $\dot{\delta\alpha}/\alpha \leq 3.1 \times 10^{-17} \text{ yr}^{-1}$	RZ2, Reactor spectrum for fresh core, 725 \pm 55 K
Cosmophysical and laboratory data			
IUCAA, Pune, India	H. Chand, R. Srianand <i>et al.</i> , 2004 [68,69]	$\delta\alpha/\alpha \leq (-60 \pm 60) \times 10^{-8}$ $\dot{\delta\alpha}/\alpha \leq 12 \times 10^{-17} \text{ yr}^{-1}$	Cosmophysical multidoublet method
Observ. de Paris France	S. Bize <i>et al.</i> 2004 [71]	$\dot{\delta\alpha}/\alpha \leq (-5 \pm 53) \times 10^{-17} \text{ yr}^{-1}$	Method of Atomic Fountains

and

$$-5.3 \times 10^{-17} \leq \overline{\delta\alpha}/\alpha \leq 1.0 \times 10^{-17} \text{ yr}^{-1}. \quad (49)$$

Thus in this case, too, we do not find with certainty a nonzero deviation of the change of α .

The results on a possible change of α based on the analysis of the cross section of ^{149}Sm in the Oklo reactor are summarized in Table XII. For comparison, we have included the cosmological results (Fig. 13) [68–70] and the results of laboratory measurements [71].

All results show that there are no grounds for an assertion that the e.m. constant has changed in the distant past. However, there is a possibility that this conclusion will be revised when the fuel burn-up is taken into account.

In the past, not only the fine structure constant could have changed, but also other fundamental constants. A review of possible changes of the fundamental constants (both experimental and theoretical) was recently published by Uzan [57]. In the case of the strong interactions described by a dimensionless theory with a running coupling constant (QCD), it is better to consider not a change of the coupling constant itself but rather changes of dimensionless ratios of quark current masses to the cutoff, $m_{u,d,s}/\Lambda$ [57]. Recently, a number of interesting papers were published [72–74] which found the limits on the change $m_{u,d,s}/\Lambda$, some of them made use of the Oklo data. According to [73], our new limits on the position of the resonance imply that the change of X_s from the time when Oklo reactor was working is restricted to

$$-4.4 \times 10^{-10} < \delta X_s/X_s < 3.7 \times 10^{-10}. \quad (50)$$

However, the discussion of this question lies outside the scope of the present paper.

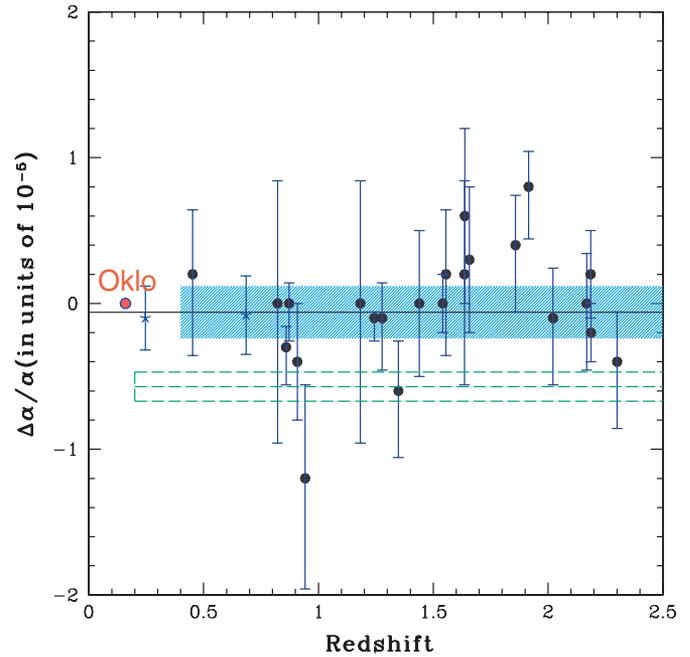


FIG. 13. (Color online) Data of Srianand *et al.* (filled circles) are plotted against the redshift [69]. Each point is the best fitted value obtained for individual systems using χ^2 minimization. The open circle is measurement from the Oklo reactor. The weighted average and 1σ range measured by Murphy *et al.* [70] are shown with horizontal dashed lines. Most of Srianand *et al.* measurements are inconsistent with this range. The shaded region represents the weighted average and its 3σ error: $\langle \delta\alpha/\alpha \rangle_w = (-60 \pm 60) \times 10^{-8}$. Within 3σ there is no variation of fundamental constants within the limits: $-25 \times 10^{-17} \leq (\Delta\alpha/\alpha \Delta t) \leq 12 \times 10^{-17} \text{ yr}^{-1}$.

IV. CONCLUSIONS

We have built a complete computer model of the Oklo reactor core *RZ2*. With the aid of present-day computational codes we have calculated in all detail the core parameters. The simulations were done for three fresh cores of different contents of uranium and water. We have also calculated the neutron flux and its spatial and energy distributions. For the three cores, we have estimated the temperature and void effects in the reactor. As expected, the neutron reactor spectrum is significantly different from the ideal Maxwell distribution that had been used by other authors to determine the cross section of $^{149}_{62}\text{Sm}$. The reactor cross section and the curves of its dependence on the shift of the resonance position ΔE_r (as a result of a possible change of fundamental constants) differ appreciably from earlier results. The effect of an influence of the reactor spectrum on the cross section of $^{149}_{62}\text{Sm}$ can be considered to be firmly established. We have studied the limits of the variation of this effect depending on the initial composition and the size of the core. The fresh bare core *RZ2* is critical for $T_C = 725 \pm 55$ K. At these temperatures, the curves of $\hat{\sigma}_{\gamma, \text{Sm}}(T_C, \Delta E_r)$ lie appreciably lower than for a Maxwell distribution. Possible values of ΔE_r lie in the range of $-73 \leq \Delta E_r \leq 62$ meV. These limits are 1.5 times more accurate than those of Dyson and Damour. For the rate of change of the e.m. constant, we find $-3.7 \times 10^{-17} \leq \overline{\delta\alpha}/\alpha \leq +3.1 \times$

10^{-17} yr^{-1} . Within these limits, we have $\overline{\delta\alpha}/\alpha = 0$. The analysis of all previous studies shows that none of them has reliably shown up a deviation from zero of the rate of change of the e.m. constant α . Because of difficulties with the detailed calculation of the burning up in large reactors, which requires a huge accumulation of statistics, we have not determined the effect of the burn-up on the neutron spectrum and on the $^{149}_{62}\text{Sm}$ cross section. Calculations of the influence of burn-up on the temperature of the active core and on the neutron spectrum are in progress.

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