OKLO REACTORS AND IMPLICATIONS FOR NUCLEAR SCIENCE

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Abstract

We summarize the nuclear physics interests in the Oklo natural nuclear reactors, focusing particularly on developments over the past two decades. Modeling of the reactors has become increasingly sophisticated, employing Monte Carlo simulations with realistic geometries and materials that can generate both the thermal and epithermal fractions. The water content and the temperatures of the reactors have been uncertain parameters. We discuss recent work pointing to lower temperatures than earlier assumed. Nuclear cross sections are input to all Oklo modeling and we discuss a parameter, the $^{175}\text{Lu}$ ground state cross section for thermal neutron capture leading to the isomer $^{176}\text{m Lu}$, that warrants further investigation. Studies of the time dependence of dimensionless fundamental constants have been a driver for much of the recent work on Oklo. We critically review neutron resonance energy shifts and their dependence on the fine structure constant $\alpha$ and the ratio $X_q = m_q/\Lambda$ (where $m_q$ is the average of the $u$ and $d$ current quark masses and $\Lambda$ is the mass scale of quantum chromodynamics). We suggest a formula for the combined sensitivity to $\alpha$ and $X_q$ that exhibits the dependence on proton number $Z$ and mass number $A$, potentially allowing quantum electrodynamic and quantum chromodynamic effects to be disentangled if a broader range of isotopic abundance data becomes available.

PACS numbers: 06.20.Jr, 07.05.Tp, 21.10.Sf, 24.30.-v, 27.20.+n, 28.20.Gd, 28.41.-i

Keywords: Oklo; natural nuclear reactors; Monte-Carlo simulation; neutron/gamma fluxes; core temperature; nuclear data; nuclear waste depository; time variation fundamental constants.
I. DISCOVERY OF THE OKLO NATURAL REACTORS

On 25 September 1972, André Giraud, Head of the French Commissariat à l’Énergie Atomique (CEA), announced the discovery of a two billion year-old nuclear reactor in Gabon, at the site of the Oklo uranium mines. The sequence of events that led to this startling announcement had begun earlier, in June 1972, at the Pierrelatte uranium enrichment plant with the observation of a small but definite anomaly in the uranium isotopic ratio for a UF₆ sample. The supply of anomalous uranium was soon traced to uranium rich ores (up to 60%) for which investigations revealed local uraninite deposits with ²³⁵U isotopic abundance of 0.600%, instead of the normal 0.7202%.

At first glance one can wonder how the Oklo reactors were able to operate when it is well known that the modern light water reactors cannot work with natural uranium, requiring instead ²³⁵U enrichment of about 3.5%. Natural uranium is composed of three isotopes with abundances today¹ of 99.2744% for ²³⁸U, 0.7202% for ²³⁵U, and 0.0054% for ²³⁴U. However, the relative enrichment of ²³⁵U increases going back in geological time because the half-life of ²³⁵U is 710 Myr while that of ²³⁸U is 4.51 Gyr. For example, ²³⁵U enrichment was 1.3% 700 Myr ago, 2.3% 1.40 Gyr ago, 4.0% 2.10 Gyr ago, and up to 17% at the time of creation of the solar system.

As reviewed by Zetterström [1], the stabilization of the Oklo area geological basement happened not earlier than 2.7 Gyr ago and the geological age of the Francevillian sediments is estimated [2] to be about 2.265 ± 0.15 Gyr. An independent constraint on the age of the Oklo phenomenon is provided by the Great Oxidation Event [3, 4]. This happened about 2.2 Gyr ago when, due to the biological activity of cyanobacteria, the oxygen content in the atmosphere of Earth increased by about a factor of a hundred. This allowed uranium to be converted from its insoluble uranium(IV) form to its soluble uranium(VI) form. Deposition of high grade uranium ores in sediments subsequently occurred when this soluble uranium was precipitated out, either by reduction back to the insoluble form (by carbon, methane or iron), or by direct microbial induced action.

Interest in natural nuclear reactors preceded their discovery by almost two decades. As early as 1953, Wetherhill and Ingram had found evidence in Congo pitchblende Xe isotopic data that, besides spontaneous fission, neutron induced fission had taken place [5].

¹ Abundance and lifetime data are from the National Nuclear Data Center (www.nndc.bnl.gov).
stated that “the deposit was twenty-five percent of the way to becoming a pile; it is interesting to extrapolate back 2000 million years . . . Certainly such a deposit would be close to being an operating pile.”

Following this suggestion, while considering the Johanngeorgenstadt (Saxony) pitchblende – a uranium ore with a minimal content of rare earth poisons, Kuroda [6] applied Fermi’s four-factor pile theory [7] and obtained estimates of neutron multiplication factors greater than unity for proper amounts of water in pitchblende. His conclusion was that “critical uranium chain reactions could have taken place if the size of the assemblage was greater than, say, a thickness of a few feet.” Kuroda looked for possible changes in the chemical composition of samples from uranium ores from several locations (but not Oklo, of course), and found no signs of chain reactions. At the time his paper was written (1956), it seemed highly unlikely that natural reactors would be found on Earth.

The first research reports on Oklo data appeared in the fall of 1972 (Bodu et al. [8], Neuilly et al. [9] and Baudin et al. [10]) and starting in 1973, the CEA launched the project “Franceville”, named after the town Franceville in the vicinity the Oklo mines. Uranium mining was suspended for two years to probe the terrain. Six natural reactor zones were discovered and samples were shared widely with the cooperation of the International Atomic Energy Agency (IAEA). In June 1975, the first international Oklo meeting took place in Libreville, with the proceedings published by the IAEA [11]. To continue the work, the IAEA and CEA established an International Working Group on Natural Reactors with a technical committee of experts. This group met in Paris in December 1977 to review progress, and published further proceedings in 1978 [12]. A review of all work done until 1990 can be found in an excellent book by Naudet [13], who was the Franceville project head.

New zones were later identified [14], and a European research program “Oklo - natural analogue for a radioactive waste repository” was initiated to study analogies between the behavior of materials in Oklo and in planned nuclear waste repositories [15]. This program was financed by the European Commission of the European Union and implemented in co-operation with institutions from other countries.

Studies of Oklo continue unabated with about 140 papers in the published literature since 2000. Recent papers split evenly between interest in the fascinating geology and operation of the reactors, and interest in what the isotopic remains can say about time variation of fundamental constants over the last two billion years. Notable earlier reviews include those
by Naudet [16], Cowan [17], Petrov [18], Kuroda [19], Meshik [20], and Barrè [21]. We use the basic information from these reviews and focus here on recent results on modeling the reactors and their implications for refining bounds on the time variation of dimensionless fundamental constants such as the electromagnetic fine structure constant.

II. OKLO REACTORS

A. The fifteen reactor zones

According to Gauthier-Lafaye [22], the chief geologist of the project “Franceville”, fourteen reactor zones were located at the Oklo-Okelóbondø area and one zone at Bangombé, 30 km away. Other authors [23] cite the total number as seventeen. With regard to the geochemical behavior of the reaction products, the zones are classified according to whether they were mined close to the surface in open pits (down to about 100 m), or mined underground at greater depth [23]. Zones of the first type (from No. 1 to 9) were certainly affected by weathering processes, while zones of the second type were weathered only little, if at all. Of these latter zones, reactor zone 10 located at a depth of 400 m is considered to be best preserved from post-reaction alterations. Located within a ground water discharge area, and being besides very shallow (10 – 12 m deep), the Bangombé reactor was in large part washed out. Regrettably, only the Bangombé Site has been preserved [24]. All Oklo-Okelóbondø zones were mined out [22].

B. Oklo isotopic data

Prior to excavation, the reactor zones contained gangue, $^{238}$U, depleted $^{235}$U, and stable fission products. These products were mostly isotopes of rare earth elements (REEs) resulting from the heavy fission fragments, and some elements, such as Zr and Y, resulting from the light fission fragments. As an example, we show in Table 1 the data for Nd, U, Ce and Sm taken from Ref. [9]. Most of the data for Nd and Ce resemble fission yields more closely than natural abundance data. The few differences are explained in what follows.

The isotope $^{142}$Nd is not produced by fission and therefore its presence in Oklo ores is due to natural Nd. This enables one to make corrections for elements present in the uranium sediments before the chain reaction took place. In addition, the elemental abundances and
isotopic concentrations in Oklo samples are influenced by neutron capture and radioactive decay. Neodymium concentrations are perturbed by the large neutron capture cross sections of $^{143}$Nd and $^{145}$Nd. When all the corrections are made, the concentrations of the neodymium isotopes correspond precisely to the fission data, and traditionally have provided the best estimates of both the neutron fluence and the total number of fissions of $^{235}$U that occurred in reactor zones.\footnote{Lanthanum isotopic data can, in principle, also give estimates of the neutron fluence\cite{25}, but, in practice, such estimates are limited by the lack of isotopic data, and are also more susceptible to uncertainties due to the variability in natural elemental abundance.} The strong deviation in the Sm data is evidently due to burning of $^{149}$Sm during reactor operation. We will discuss the $^{149}$Sm data in detail later; this is the channel of greatest interest in studies of the time evolution of fundamental constants.

Since 1972 similar data have been obtained by mass spectrometry methods for many elements. Isotopic composition and elemental abundances have been reviewed in Ref.\cite{13} for reactor zones 2 and 3, in Ref.\cite{23} for reactor zone 9, and in Ref.\cite{26} for reactor zone 10. In an interesting development, the isotopic enrichment of the gaseous fission products Kr and Xe, trapped in minerals, was studied\cite{27–29}, with important results which we will discuss in the next subsection. One can deduce from the totality of all these data the neutron fluence, the average power of the reactors, the duration of reactor operation and the age of the Oklo phenomenon (i.e. how long ago it occurred). While some author prefer more recent times of occurrence, the most accurate determination is reported\cite{30,31} as 1950±40 Myr, a result in good agreement with the geological age mentioned above. Results for the duration of reactor operation for Oklo zones 10 and 13 and the Bangombe site range from 100 to 300 thousand years\cite{26}.

\section*{C. Periodic mode of operation}

In most of the earlier work on the Oklo phenomenon, a steady-state mode of operation was tacitly accepted. With this assumption, a neutron fluence in a single zone is of the order of $10^{21}$ cm$^{-2}$ (see, for example, Ref.\cite{26}), and an operation duration of about $3 \times 10^5$ yr leads to an average neutron flux density of $10^8$ cm$^{-2}$s$^{-1}$, about five orders of magnitude less than present day power reactors. In a pulsed mode of reactor operation, however, the instantaneous flux can be much higher.

In 1977, Yu. Petrov\cite{32} suggested a self-regulating mechanism for pulsed operation at...
TABLE I. Isotopic data of the ore sample M from the Oklo mine compared with natural concentrations and with cumulative fission yields [Neuilly et al., C. R. Acad. Sci. Paris, Ser. D 237 (1972) 1847].

| Isotopes | Natural concentration | $^{235}$U fission Oklo M |
|----------|-----------------------|-------------------------|
| $^{142}$Nd | 27.11 % | 0.00 % | 1.38 % |
| $^{143}$Nd | 12.17 % | 28.8 % | 22.1 % |
| $^{144}$Nd | 23.85 % | 26.5 % | 32.0 % |
| $^{145}$Nd | 8.30 % | 18.9 % | 17.7 % |
| $^{146}$Nd | 17.22 % | 14.4 % | 15.6 % |
| $^{148}$Nd | 5.73 % | 8.26 % | 8.01 % |
| $^{150}$Nd | 5.62 % | 3.12 % | 3.40 % |
| $^{235}$U | 0.720 % | 0.440 % |
| $^{140}$Ce/$^{142}$Ce | 7.99 % | 1.06 % | 1.57 % |
| $^{149}$Sm/$^{147}$Sm | 0.92 % | 0.47 % | 0.003 % |

*Isotopic concentrations are in atomic percentages. The relative precision is about 0.2% for the natural abundances, 0.5% for fission data, and from 2% to 3% for Oklo data.

Oklo based on the negative void temperature coefficient. According to this mechanism, with increasing reactor temperature, neutron moderating water is converted into steam which can even leave the active core. As a result, the reactor shuts down, cools off, and only restarts when water seeps back into the reactor zone again.

The time scale for such operation remained uncertain until 1990 when Kuroda [33, 34] came up with the idea of estimating the cooling period by analyzing data [27] on the anomalous isotopic composition of Oklo xenon. In particular he compared the excesses (relative to standard cumulative fission yields) of the isotopic ratios Xe(132/136) and Xe(134/136) and took note of the fact that none of these Xe isotopes is a direct result of fission. Their cumulative yields are due to fission fragment precursors having quite different half-lifes. Kuroda explained the excess for Xe(132/136) by taking into consideration the different half-lifes of the precursors, $^{132}$Te (78.2 h) and $^{134}$Te (0.7 h). Assuming a pulsed mode of operation with a reactor-on time, $\Delta t_{on}$, much larger than the reactor-off time, $\Delta t_{off}$, he found 2.5 h
< $\Delta t_{\text{off}} < 3 \text{ h}$. The Xe data of Ref. [27] were for the uranium grains taken directly from several active Oklo cores.

In the years 2000 – 2004, Meshik et al. [28, 29] studied the Xe isotopic anomalies in Oklo zone 13, both in uranium grains as well as in Al phosphates formed through the action of heated water. They found that Xe was trapped preferentially in the phosphates and its concentration exceeded that in uranium by several orders of magnitude. The resulting better sensitivity of the isotopic analysis allowed them to consider the influence of many precursors in the mass chains leading to the stable Xe isotopes. They were able to deduce values of $\Delta t_{\text{off}} = 2.5$ hours and $\Delta t_{\text{on}} = 0.5$ hours. Although these results apply strictly only to RZ13, we take the numbers to be typical for all the reactor zones, and use them in one of the following sections.

III. MONTE-CARLO SIMULATIONS OF OKLO REACTORS

To date, the Oklo uranium deposits are the only known place on Earth where two billion years ago all conditions were appropriate for fission chain reactions. Those conditions are: (i) a high concentration of uranium in different zones, with the $^{235}$U abundance being at least 3%; (ii) a large amount of water in the pores and cracks of minerals, and; (iii) the absence or near-absence of neutron poisons such as boron, lithium and the REEs. Elemental compositions and geometries of six Oklo zones were already reported at the first Oklo symposium [11]. These data were enough to apply theories of neutron chain reactors [35] to neutronic calculations for multiplication factors, neutron fluxes and other reactor physics parameters. Naudet and Filip developed the multigroup code BINOCULAR [36], which solved the neutron transport equations for some Oklo zones, especially the largest RZ2. Interest in radioactive waste storage lead to the development of sophisticated deterministic codes for modeling criticality and these have also been applied to Oklo [37, 38].

Most recently, a probabilistic approach based on the application of the code MCNP [39] has been utilized. The MCNP code models neutron transport using Monte Carlo methods and takes advantage of continuous libraries of energy dependent neutron cross sections. It can deal with practically any realistic geometry of an active reactor core. Modeling with MNCP has been performed for zones RZ2 [40, 41], RZ3 [42], RZ9 [43, 44], and RZ10 [41]. We illustrate some details of such modeling using the examples of zones RZ2 and RZ10.
A. Active core composition and criticality

The MCNP code input for any Oklo reactor consists of a description of its geometry with relevant physical dimensions, the operating temperature, and specification of the amounts of uraninite (a variety of UO$_2$), gangue and water. Typically, reactor zones are lens shaped layers, about 10 m long, about 10 m wide and up to 0.8 m thick. The uraninite content varies from zone to zone reaching as high as 80wt.% in some samples. The gangue is composed of clays – the hydrated silica-aluminates of different metals. Data for several zones are detailed in Ref. [13]. The amount of water during the time of reactor operation remains very uncertain. Water, however, defines the shape of the neutron spectrum and can therefore be deduced by varying its amount until the spectral indices$^3$ of the calculated neutron flux coincide with the indices deduced from the isotopic data. The output of MCNP consists of the criticality value and the energy dependence of the neutron spectrum.

In Ref. [41], the geometries of Oklo zones RZ2 and RZ10 were approximated as flat cylinders of 6 m diameter and 70 cm height, surrounded by a 1 m thick reflector consisting of water saturated sandstone. The compositions (in g/cm$^3$) of the two reactors are shown in Table 2. The composition of RZ2 is from Ref. [13] and that of RZ10 from Refs. [14] and [26]. The total density of the active core material at ancient times was about 4 g/cm$^3$ for RZ2 and 3 g/cm$^3$ for RZ10. The most striking difference between RZ2 and RZ10 is the rather small, 28wt.% content of uraninite UO$_2$ in RZ10. As a result, RZ10 cannot become critical with a poison more than 1 ppm of $^{10}$B equivalent while RZ2 can accommodate 10 ppm of $^{10}$B equivalent. The amount of water (H$_2$O) shown in Table 2 is the total amount, including

$^3$ Spectral indices are a measure of the epithermal neutron fraction in the neutron spectrum.
B. Neutron and $\gamma$-ray fluxes

The MCNP code allows one to model both the neutron fluxes in reactors and $\gamma$-ray fluxes. Because the temperature in the active Oklo cores remains uncertain (see later discussion), Ref. [41] obtained the neutron fluxes of zones RZ2 and RZ10 for a set of different temperatures: 20 $^\circ$C, 100 $^\circ$C, 200 $^\circ$C, 300 $^\circ$C, 400 $^\circ$C, and 500 $^\circ$C. The calculations of the energy dependent neutron fluxes were performed with energy bins on a lethargy grid, where the (dimensionless) lethargy $u$ ran from 23 to 9.3 in steps of 0.1. This gave neutron energies $E = (10^7 \text{eV}) e^{-u}$ from about 1 meV up to about 1 keV, and bin widths $\Delta E \approx 0.1E$. In Fig. 1, we show the modeled spectra as a function of neutron energy $E$ for RZ10. The neutron fluxes are the family of curves starting at the lower left of the figure. The leftmost curve corresponds to a temperature of 20 $^\circ$C and the rightmost curve to 500 $^\circ$C. Uranium absorption resonances are prominent in the epithermal region.

For some purposes, the neutron density is a more relevant than the neutron flux. The neutron flux $\phi(E)$ is related to the neutron density $n(E)$ by $\phi(E) = n(E) v(E)$, where $v(E)$ is neutron velocity. Neutron densities (normalized to one neutron per unit volume) are the families of curves starting upper left in Fig. 1. The topmost curve corresponds to 20 $^\circ$C and the lowest curve corresponds to 500 $^\circ$C. These spectra demonstrate clearly the presence of two components in two different energy regions: the thermal neutron region below about 0.5 – 1.0 eV and the so-called epithermal or $1/E$ region.

Despite the fact that the MCNP spectra do not exactly follow $1/E$-behaviour in the epithermal region, Oklo spectral indices $r_O$ can be deduced from flux plots. Figure 2 illustrates the methodology and values determined from Ref. [41] for $r_O$ are listed in Table 2. They agree with “measured” values deduced from analysis of concentrations of $^{235}$U and of the fission products $^{143}$Nd and $^{147}$Sm [13, 14, 26, 30]: $r_O = 0.20 - 0.25$ for RZ2 and $r_O = 0.15 \pm 0.02$ for RZ10.

Gamma-ray fluxes can, in principle, contribute to post-processing of fission products. The issue of whether this was a concern for Oklo was investigated for the first time in Ref. [45]. Gamma-rays in a reactor arise in three ways:

- prompt $\gamma$-ray emission during fission events (eight $\gamma$-rays per fission, with a total
FIG. 1. Reactor zone RZ10 MCNP neutron fluxes and neutron densities for different temperatures [C. R. Gould, E. I. Sharapov, and S. K. Lamoreaux Phys. Rev. C 74 (2006) 24607]. The neutron fluxes are plotted as $E \phi(E)$ and are the family of curves starting from a temperature of 20 °C at the lower left. The neutron densities are the families of curves starting upper left.

- Energy of about 7.0 MeV,

- Prompt $\gamma$-ray emission following neutron radiative capture in materials of the reactor (also with a total energy of about 7.0 MeV)

- Delayed $\gamma$-ray emission from the decay of fission products (a total energy of about 6.3 MeV).

Prompt $\gamma$-production was modeled for the Oklo reactor zone RZ10 with the MCNP code using the same input files as for the neutron flux modeling. The modeling was performed for the pulsed mode of the reactor operation discussed above (reactor on for 0.5 h and off for 2.5 h). Delayed $\gamma$-production as a function of time was studied analytically by Way [46] and later numerically with improved decay data libraries [47]. The results of Ref. [45] are shown in Fig. 3. As might be expected, the total prompt $\gamma$-ray flux dominates and is about $3 \times 10^9$
FIG. 2. Realistic RZ10 MCNP neutron flux at thermal energies compared to model fluxes with different Oklo spectral indices $r_O$.

$\gamma$ cm$^{-2}$ s$^{-1}$. However, at some energies, the delayed $\gamma$-ray flux is significant. Implications of these calculations for lutetium thermometry in RZ10 will be discussed in the next section.

IV. REACTOR TEMPERATURES

Much attention has been focused on the temperatures at which the Oklo reactors operated. This parameter sets the shape of the energy spectrum, an important input to Oklo analyses investigating possible time variations in fundamental constants. The pulsed mode of operation currently favored indicates no single temperature can be appropriate. Nevertheless an average temperature is crucial to establishing general characteristics, and will be an appropriate parameter as long as the system is responding in a linear fashion.

The reactors likely operated under conditions similar to present day PWR systems, with pressures about 150 atm and temperatures of about 300 °C. The critical point for water (no liquid possible at any pressure) occurs at about 374 °C and 218 atm. This sets a plausible upper limit to the temperature although some modeling of Oklo reactors has achieved criticality at higher temperatures, even up to about 550 °C in some cases [40].
FIG. 3. Prompt and delayed $\gamma$-ray fluxes in Oklo zone RZ10. The fluxes are calculated for an 18 kW reactor, on for 0.5 hr and off for 2.5 hr. The prompt flux is the upper line. The lower line is the delayed flux, taking into account the equilibrium flux associated with N previous reactor-on pulses ($N \gg 1$).

Lutetium thermometry first proposed by Wescott [48] can in principle provide a direct measure. The method depends on the overlap of the thermal neutron spectrum with a low energy neutron capture resonance, in this case at 141 meV in $^{176}$Lu. The temperature is extracted by comparing the $^{176}$Lu/$^{175}$Lu ratio and noting the depletion of $^{176}$Lu will be strongly dependent on temperature whereas the depletion of $^{175}$Lu is not. Effective cross sections for the two isotopes are listed in Table III.

The method was first applied to Oklo studies by Holliger and Devillers [49] who found temperatures of $T_O = 260 \, ^{\circ}C$ and $280 \, ^{\circ}C$ for RZ2 and RZ3, respectively. The later more realistic modeling of Onegin [42] found $T_O = (182 \pm 80) \, ^{\circ}C$ for RZ3. However for RZ10, one of the most well characterized zones, Hidaka and Holliger [26] succeeded in getting a result ($T_O = 380 \, ^{\circ}C$) for only one sample. Three other samples yielded only an anomalously high bound, $T_O > 1000 \, ^{\circ}C$.

Reasons for these problematic results were investigated by Gould and Sharapov [50]. They noted the dependence on a potentially uncertain parameter, $B^g$, the $^{175}$Lu ground state to isomer state capture branching ratio. This issue emphasizes anew the role of nuclear data in understanding the Oklo phenomenon, a point which was stressed already [51] at the beginning of studies of Oklo. Since then most of the nuclear data required for Oklo
TABLE III. Lutetium effective cross sections $\hat{\sigma}_5$ and $\hat{\sigma}_6$ for the Oklo RZ10 reactor at temperatures $T_O$ from 0 °C to 600 °C. (C. R. Gould and E. I. Sharapov, Phys. Rev. C 85 (2012) 024610).

| $T_O$ (°C) | $\hat{\sigma}_5$ (kb) | $\hat{\sigma}_6$ (kb) |
|------------|------------------------|------------------------|
| 0          | 0.115                  | 4.216                  |
| 20         | 0.115                  | 4.487                  |
| 100        | 0.115                  | 5.359                  |
| 200        | 0.115                  | 6.310                  |
| 300        | 0.114                  | 7.013                  |
| 400        | 0.114                  | 7.544                  |
| 500        | 0.114                  | 7.715                  |
| 600        | 0.114                  | 7.750                  |

analyses has been improved considerably, in particular the mass distributions of the fission products of $^{235}$U, $^{238}$U and $^{239}$Pu, the isotopic ratio data Nd/$^{238}$U and Bi/$^{238}$U, the Sm, Gd and Nd isotopic ratios, and various decay constants. Nevertheless, it appears that the lutetium branching ratio $B^g$ for thermal neutrons remains very uncertain. This ratio has been the subject of intensive study by the astrophysics community because of its importance in understanding the s-process in stars. New measurements [52, 53] at 5 and 25 keV gave values for $B^g$ significantly different from the thermal energy value which is implied by the neutron cross sections in Mughabghab’s *Atlas of Neutron Resonances* [54] and needed for thermometry. Using the most accurate of these improved “astrophysical” values, the metasample reanalysis for RZ10 yields $T_O = (100 \pm 30)$ °C, much lower than the earlier values cited. The Onegin RZ3 result will also change if a lower value of $B^g$ is used, being instead closer to about 100 °C. However, because of the presence of p-wave neutrons at keV energies (which are absent at thermal energies), the $B^g$ values of Refs. [52] and [53] may not be directly applicable to thermal neutrons. A precision measurement of $B^g$ at thermal energies would clearly help to resolve this issue and ways of performing this experiment have been pointed out in Ref. [55].

An alternate explanation for problems with lutetium thermometry, burn up of $^{176}$Lu in the $\gamma$-ray flux of the reactors, was investigated in Ref. [45]. Again, improved nuclear data is required, this time for the integral cross sections [56, 57] for photo-excitation of the
specific excited states of $^{176}$Lu which decay to metastable $^{176m}$Lu. Excitation of this isomeric state (which then beta decays to $^{176}$Hf) is an important loss mechanism in astrophysical environments [58]. However, the $\gamma$-ray fluxes in Oklo reactors obtained in Ref. [45] prove to be many orders of magnitude too low for these ($\gamma$, $\gamma'$)-reactions to impact application of lutetium thermometry.

In summary, despite uncertainties, most of the later Oklo analyses together with the latest nuclear data favor lower temperatures compared to earlier work.

V. GEOLOGICAL REPOSITORY OF NUCLEAR WASTES

Oklo is unique not only for being the site of multiple natural reactors, but also as a place where fission products have been preserved undisturbed for over two billion years. The zones therefore present a unique opportunity for learning more about the long-term geochemical behavior of radioactive wastes from nuclear reactors. This was realized from the very beginning by US scientists who accordingly started an Oklo program relevant to waste management technology [59].

The chief concern of this program was determination of the relative mobilities of the reactor products in the Oklo environment and the migration/retention behavior of various elements. One of the most important, and surprising, early findings was that uranium and most of the REEs did not experience significant mobilization in the past two billion years. Results have been published in proceedings of conferences [11, 12] and reviewed in some detail in Ref. [59]. Further work followed in a European Commission Program “Oklo – natural analogue for a radioactive waste repository”. The focus here was on the considerably more complicated situation involving storage of waste materials from modern high power reactors. Results obtained from this program can be found in Refs. [15, 60] and [61]. Modeling the potential for criticality in geological storage is an important issue and continues to be studied intensively. Here again the Oklo reactor zones have served as a testing ground for new approaches and new computation codes [37, 38, 62, 63].

Because the wastes were contained successfully in Oklo, it appears not unrealistic to hope that long term disposal in specially selected and engineered geological repositories can be successful [64].
VI. STABILITY OF THE FINE STRUCTURE CONSTANT AND LIGHT QUARK MASSES

The late Alexander Shlyakhter was the first to recognize the potential of Oklo neutron absorption cross sections to constrain fluctuations in fundamental constants [65]. He seems to have derived his inspiration from an appealing analogy with the ordinary radio receiver (to which he alludes): just as the reception frequency of a radio is altered when the parameters of its tuning circuitry are adjusted so the energy of a neutron capture resonance shifts as “constants” in the nuclear Hamiltonian change. Armed with the confidence of a pioneer who knows there is no earlier explorer to contradict him, Shlyakhter presented crude order of magnitude estimates to demonstrate the sensitivity of resonance shifts (expounded with a few short sentences). He deduced upper bounds on the time-dependent variation of the strong, weak and electromagnetic coupling constants, which were significantly better than previous upper bounds due to Dyson [66, 67] and Davies [68].

The intervening years have witnessed a number of attempts to make more convincing estimates with the Oklo energy shift data [69–81]. Some of this work has been driven by the realization that, within many theories beyond the standard models of particle physics and cosmology, fundamental constants are, in effect, dynamical variables [82, 83] (see also the reviews in Refs. [84] and [85]). More pressing, perhaps, have been the empirical findings [86–90] suggesting a change in the fine structure constant $\alpha$ over cosmological time scales. As emphasized in Refs. [91] and [92], grand unification schemes automatically imply that other fundamental Standard Model parameters should evolve with time. Relating the Oklo phenomenon to the behaviour of parameters in quantum chromodynamics (QCD) has been frustrated by the difficulties of the confinement problem compounded by the usual uncertainties of the nuclear many-body problem. An elegant scheme for extracting information on the fine structure constant $\alpha$ of quantum electrodynamics (QED) has, however, been developed [72]. In Refs. [70] and [72], the implications of Oklo for the weak interaction (specifically, the Fermi coupling constant $G_F$) are considered briefly, but it would seem reasonable to ignore them (as other authors have done): in the absence of parity-violation, electromagnetic and QCD effects mask any contribution of the weak interaction to neutron capture.

Despite the appeal in Ref. [91] to obtain “more accurately the constraints that [the Oklo reactor] imposes on the space of coupling constants”, most analyses of Oklo restrict
themselves to inferring the corresponding limits on the change in $\alpha$ only. The 97.3 meV resonance seen in neutron capture by $^{149}\text{Sm}$ (which we term the Sm resonance below) has received most attention.

A. Sensitivity of an energy shift to the fine structure constant

The analysis of Damour and Dyson in Ref. [72] begins with the approximation of the neutron capture resonance energy $E_r$ as a difference in expectation values of the nuclear Hamiltonian $\hat{H}$:

$$E_r = \langle r | \hat{H} | r \rangle - \langle g | \hat{H} | g \rangle,$$

(1)

where $|g\rangle$ denotes the eigenket of the ground state of the target nucleus and $|r\rangle$ denotes the eigenket of the compound nucleus state formed by the capture of the neutron. By the Feynman-Hellmann theorem, the rate of change of $E_r$ with $\alpha$ is

$$\frac{dE_r}{d\alpha} = \langle r | \frac{\partial \hat{H}}{\partial \alpha} | r \rangle - \langle g | \frac{\partial \hat{H}}{\partial \alpha} | g \rangle.$$

If, as in Ref. [72], small electromagnetic effects (like magnetic-moment interactions and QED radiative corrections to nucleon properties) are neglected, then only the Coulomb potential energy operator $\hat{V}_C$ (proportional to $\alpha$) contributes to the partial derivative of $\hat{H}$ — i.e.

$$\alpha \frac{dE_r}{d\alpha} = \langle r | \hat{V}_C | r \rangle - \langle g | \hat{V}_C | g \rangle.$$

(2)

To evaluate the difference in Coulomb energies in Eq. (2), Damour and Dyson resort to replacing the expectation values by the classical Coulomb energies

$$\frac{1}{2} \int V_r \rho_r \, d^3r,$$

where $V_i$ is the electrostatic potential associated with the charge density $\rho_i$. With this approximation, Eq. (2) becomes

$$\alpha \frac{dE_r}{d\alpha} \simeq \frac{1}{2} \int (V_r \rho_r - V_g \rho_g) \, d^3r.$$

(3)

The ingenious step of Damour and Dyson is to now use Poisson’s equation along with Green’s second identity and recast the right-hand side of Eq. (3) in the form

$$\int V_r \delta \rho \, d^3r - \frac{1}{2} \int \delta V \delta \rho \, d^3r$$

(4)
in which $\delta \rho = \rho_r - \rho_g$ and $\delta V = V_r - V_g$.

Equation (4) permits two simplifications. The integral in the second term is positive because it is twice an electrostatic self-energy. For the purposes of establishing an upper bound on $dE_r/d\alpha$, the second term in Eq. (4) can be discarded. The second more subtle advantage of Eq. (4) is that one can exploit the fact that any effects of deformation (either static or dynamic) should be less pertinent to the excited compound nucleus state $|r\rangle$ than they are to the ground state $|g\rangle$. Damour and Dyson adopt for $V_r$ the potential $V_u$ of a uniformly charged sphere of radius $R_r$ and charge $Q = Ze$ which they claim implies that
\[
\int V_r \delta \rho \, d^3r \approx \int V_u \delta \rho \, d^3r = -\frac{(Ze)^2}{2R_r^2} \left[ r^2 \right]_{rg},
\]
where
\[
\left[ r^2 \right]_{ij} \equiv \frac{1}{Ze} \int r^2 (\rho_i - \rho_j) \, d^3r.
\]
Determination of an upper bound on $dE_r/d\alpha$ is reduced to finding a positive lower bound on $[r^2]_{rg}$.

To this end, it suffices to make the reasonable supposition that the charge distribution is more diffuse in the excited state $|r\rangle$ than in the ground state $|\tilde{g}\rangle$ of the same nucleus or that $[r^2]_{rg} \geq 0$. It follows that
\[
\left[ r^2 \right]_{rg} = \left[ r^2 \right]_{\tilde{g}g} + \left[ r^2 \right]_{\tilde{g}g} \geq \left[ r^2 \right]_{\tilde{g}g},
\]
where, significantly, the (positive) numerical value of $[r^2]_{\tilde{g}g}$ is obtained directly from experimental isotope-shifts [93] (see Ref. [72] for details). Finally, one obtains the inequality
\[
\alpha \frac{dE_r}{d\alpha} < -\frac{(Ze)^2}{2R_r^2} \left[ r^2 \right]_{\tilde{g}g},
\]
which is the result Damour and Dyson use in their interpretation of Oklo data.

The analysis of Damour and Dyson leading to Eq. (6) has been subject to some criticisms, most trenchantly in Ref. [73]. Remarkably, there is one implicit and unfounded assumption that seems to have gone unnoticed. There are also a number of nuclear structure issues which are not addressed in Ref. [72] or elsewhere. We discuss these matters, before taking up the comments of Ref. [73].

In their derivation of Eq. (5), Damour and Dyson take the potential $V_u$ of the uniformly charged sphere to be
\[
V_u = V_{\text{inside}} = \frac{Ze}{2R_r} \left[ 3 - \left( \frac{r}{R_r} \right)^2 \right]
\]
for all radial distances \( r \) (from the center of the charge distribution), whereas, in fact, \( V_u = V^{\text{inside}} \) only for \( r < R_r \), while, for \( r \geq R_r \), \( V_u = V^{\text{outside}} \equiv Z e/r \). Accordingly, Eq. (5) should, in principle, be replaced by the far less appealing result

\[
\int V_r \delta \rho \, d^3r \approx -\left( \frac{(Ze)^2}{2R_r^3} \right)_{\gamma} + \int_{R_r}^{\infty} (V^{\text{outside}} - V^{\text{inside}}) \delta \rho \, d^3r.
\]  

(7)

A back-of-envelope estimate suggests that the integral on the right-hand side of Eq. (7) is comparable to the first term (the contribution retained by Damour and Dyson). The sign of the integrand in this integral is determined by the sign of \( \delta \rho \), which is almost certainly positive for \( r \geq R_r \) (in view of the greater diffusiveness of the charge distribution of \( |r\rangle \)). As seen above, the first term on the right-hand side of Eq. (7) is negative. Thus, unfortunately, the possibility exists that there could be a significant cancellation.

The approximation of \( V_u \) by \( V^{\text{inside}} \) for all values of \( r \) is in the spirit of early studies [94] of the effect of the Coulomb interaction on nuclear properties, where the rapid drop off in nuclear wave functions was presumed to guarantee that the difference between \( V_u \) and \( V^{\text{inside}} \) for large \( r \) has negligible effect. It is now known that, for \( Z \geq 28 \), the electric field outside the nucleus contributes typically about 80% of a nuclear Coulomb energy [95].

The earlier simplification leading to Eq. (3) amounts to retaining only, in the parlance of nuclear many-body theory, the direct part of the difference in Coulomb energies. One can seek to improve upon this, in principle, by including an exchange contribution (arising from the fermion character of nucleons) and a spin-orbit term (proportional to the derivative of the electrostatic potential). The importance of such corrections can most simply be gauged with the analytic expressions for Coulomb energies of section 9 in Ref. [95]. These formulae have been inferred from a phenomenological study of Coulomb displacement energies for spherical nuclei with \( Z \geq 28 \) and contain multiplicative corrections accommodating the diffuseness of the charge distribution’s surface. A prescription for estimating the effect of deformation is also given, namely to reduce the direct Coulomb energy by a factor of \( (1 - \beta^2/4\pi) \), where \( \beta \) is the quadrupole deformation parameter. In fact, the results of Ref. [95] imply that, for the isotopes considered in Ref. [72] (and subsequent analyses of Oklo data), all the above-mentioned effects induce a decrease of less than 3% in a ground state Coulomb energy. Hence, Damour and Dyson are completely justified in neglecting them.

As regards corrections due to inter-nucleon correlations (a topic not considered explicitly in Ref. [95]), it can be argued that these should be negligible in an analysis based on Eq. (7).
For the purposes of establishing an upper bound, \([r^2]_{rg}\) is replaced by \([r^2]_{\tilde{g}g}\), which is taken from experiment and, hence, automatically includes any such corrections. Since the integral in Eq. (7) is over a region of low nucleon density, it should be insensitive to short range correlations. Long range correlations are responsible for nuclear deformation, the effect of which on Coulomb energies has already been discounted.

We can summarize our analysis as follows: for the purpose of order of magnitude estimates, it is plausible that

\[
\left| \frac{\alpha}{d\alpha} \right| \sim \frac{(Ze)^2}{2R^3} \left[ r^2 \right]_{\tilde{g}g},
\]

(8)

where \(R_r\) is the equivalent charge radius of the neutron capture resonance and \([r^2]_{\tilde{g}g}\) is the difference between the mean square charge radii of the ground states \(|\tilde{g}\rangle\) and \(|g\rangle\) (of the daughter and target nucleus, respectively). We have dropped the integral in Eq. (7) to recover what looks like the result of Damour and Dyson, but there are two differences. First, we do not believe that it is appropriate to associate confidence levels with bounds extracted using Eq. (8). Second, we have not committed ourselves to the numerical values of \(R_r\) and \([r^2]_{\tilde{g}g}\) adopted in Ref. [72] for the \(E_r = 97.3\,\text{meV}\) resonance in neutron capture on \(^{149}\text{Sm}\).

To close, we address briefly the concerns expressed in Ref. [73]. The validity of Eq. (2) is questioned on the grounds that the nuclear kinetic energy depends implicitly in \(\alpha\) (through the role of Coulomb repulsion in determining the size of a nucleus). Our presentation makes clear that Eq. (2) is a straightforward consequence of the Feynman-Hellmann theorem. For the derivative of a stationary state expectation value with respect to a parameter in the Hamiltonian, only \textit{explicit} dependence on the parameter is relevant, not any implicit dependence of the stationary state eigenfunctions. The Feynman-Hellmann theorem is, admittedly, counter-intuitive.

The other issue raised in Ref. [73] is the need to extend the analysis of Damour and Dyson to include the time dependence of parameters of the strong nuclear interaction \(\hat{V}_N\). The dependence on a (dimensionless) strength \(\alpha_s\) of \(\hat{V}_N\) is considered. The focus on \(\alpha_s\) is unfortunate because its relation to parameters of QCD is unclear (as the authors of Ref. [73] readily acknowledge). The subsequent analysis is invalidated by the omission of the contribution of the kinetic energy operator \(\hat{T}\) to the relation for \(E_r\) in terms of \(\alpha_s\) and \(\alpha\). From Eq. (1), with [96] \(\langle \langle \hat{A} \rangle \rangle = \langle r|\hat{A}|r\rangle - \langle g|\hat{A}|g\rangle\),

\[
E_r = \langle \langle \hat{T} \rangle \rangle + \langle \langle \hat{V}_N \rangle \rangle + \langle \langle \hat{V}_C \rangle \rangle,
\]
whereas Ref. [73] employs (in our notation) \( E_r = \langle \hat{V}_N \rangle + \langle \hat{V}_C \rangle \). A Fermi gas model estimate of \( \langle \hat{T} \rangle \) for the Sm resonance considered by Damour and Dyson is 23.5 MeV, an order of magnitude bigger than \( \langle \hat{V}_C \rangle \)!

(From above, \( \langle \hat{V}_C \rangle \sim -1 \text{ MeV} \).)

### B. Sensitivity of an energy shift to light quark masses

In Ref. [72], it was recognized (by appealing to the character of chiral perturbation theory) that Oklo data could, in principle, be used to place constraints on the time variation of the mass ratios \( m_l/m_p \), where \( m_l \) denotes the mass of either light (u or d) quark and \( m_p \) is the proton mass. Despite the fact that there are good reasons [97] for using \( m_p \) as the mass unit for problems involving QCD and nuclear physics in preference to the mass scale \( \Lambda \) of QCD, work subsequent to Ref. [72] has considered the dimensionless ratio \( X_q = m_q/\Lambda \), where \( m_q = \frac{1}{2}(m_u + m_d) \). The most complete analysis to date is that of Flambaum and Wiringa in Ref. [81]. There have also been several studies of the implications of Oklo for changes in \( X_s = m_s/\Lambda \) (\( m_s \) being the mass of the strange quark), but recent model independent results [98, 99] on pertinent hadronic properties (sigma terms) are discouraging. (In this subsection, all masses are in units such that \( \Lambda = 1 \).)

Central to the approach of Flambaum and Wiringa is their conjecture that the non-Coulombic contribution to the any shift in \( E_r \) is independent of mass number \( A \) (and proton number \( Z \)). More precisely, their working assumption is that this property applies to the energies of any weakly bound states as well as any resonances close to the neutron escape threshold. Arguments based on the Fermi gas model (ignoring configuration-mixing) and a small sample of quantitative results (for \( A = 6 \) to \( A = 9 \) nuclei) are given in support of this conjecture and then it is invoked to infer that

\[
m_q \frac{dE_r}{dm_q} \sim 10 \text{ MeV}
\]

from calculations in light nuclei of the kind performed in Ref. [100]. In fact, Flambaum and Wiringa compute shifts not in \( E_r \) per se but in the difference of ground states energies

\[
S \equiv \langle g | \hat{H} | g \rangle - \langle \tilde{g} | \hat{H} | \tilde{g} \rangle;
\]

which is related to \( E_r \) by \( E_r = E^* - S \), \( E^* \) being the excitation energy of the compound nucleus state \( |r\rangle \) relative to the ground state \( |\tilde{g}\rangle \) of the same nucleus. (If \( |r\rangle \) is an excited
state of the nucleus $^4_2X$, then $|g\rangle$ is the ground state of the isotope $^{A-1}_{Z^2}X$.) It is tacitly assumed in Ref. [81] that shifts in $E^* - S$ resemble those in $S$. The result in Eq. (9) is finally corroborated with an estimate based on a nuclear structure model appropriate to the study of heavy nuclei (the Walecka model).

To establish the quark mass dependence of $S$ for light nuclei, Flambaum and Wiringa employ the variational Monte Carlo method together with the Argonne $v_{18}$ (AV18) two-nucleon and Urbana IX (UIX) three-nucleon interactions [101]. They determine how ground state energies change for independent 0.1% variations in the masses $m_H$ of the hadrons $H$ deemed most important to low-energy nuclear dynamics: nucleons ($H = N$), deltas ($H = \Delta$), pions ($H = \pi$) and, lastly, a vector meson ($H = V$), which simulates the short-range interactions associated with the $\rho$ and $\omega$ mesons.\(^4\) Their results are reported as the

| TABLE IV. $K_H^E$ and $K_E$ values for $A = 5$ to $A = 9$ nuclei (AV18+UIX Hamiltonian).\(^a\) |
|-----------------|-----|-----|-----|-----|-----|-----|-----|-----|-----|
| $H$             | $^5\text{He}$ | $^6\text{He}$ | $^6\text{Li}$ | $^7\text{Li}$ | $^7\text{Be}$ | $^8\text{Be}$ | $^9\text{Be}$ |
| $N$             | 13.31 | 13.66\(^d\) | 15.78\(^d\) | 14.41 | 17.30\(^d\) | 19.34\(^b\) | 15.53 | 16.29 | 14.36 | 16.09\(^b\) |
| $\Delta$        | -10.24 | -10.54\(^d\) | -12.25\(^d\) | -10.80 | -13.48\(^d\) | -14.92\(^b\) | -11.96 | -12.56 | -11.11 | -12.39\(^b\) |
| $\pi$           | -5.82 | -5.98\(^d\) | -7.02\(^d\) | -6.31 | -7.72\(^d\) | -8.78\(^b\) | -6.91 | -7.26 | -6.31 | -7.27\(^b\) |
| $V$             | 40.87 | 42.04\(^d\) | 49.09\(^d\) | 43.48 | 54.04\(^d\) | 60.46\(^b\) | 48.11 | 50.53 | 44.40 | 50.21\(^b\) |

$K_E$

- set 1\(^c\): -1.24, -1.28\(^d\), -1.50, -1.67\(^d\), -1.93, -1.50, -1.57, -1.35, -1.59
- set 2a: 0.011, 0.013, -0.010, -0.033, -0.011, -0.080, -0.021, -0.024, 0.014, -0.054
- set 2b: 0.42, 0.43, 0.48, 0.40, 0.53, 0.52, 0.46, 0.48, 0.46, 0.45
- set 2c: 0.83, 0.85, 0.97, 0.84, 1.07, 1.13, 0.94, 0.99, 0.90, 0.95

\(^a\) Unless indicated otherwise, $K_H^E$ values are from Table IV in Ref. [100]. Sets 2a, b and c of $K_E$’s are obtained with set 2 of the $K_H^q$’s in Table V and $K_V^q = 0.6, 0.7$ and 0.8, respectively (choices motivated in the text).

\(^b\) From Table III in Ref. [81].

\(^c\) From Table I in Ref. [81].

\(^d\) R. B. Wiringa, private communication.

\(^4\) Actually, the AV18+UIX Hamiltonian only depends explicitly on the masses of the pions, the proton and the neutron. Details of how the effects of changes in $m_\Delta$ and $m_V$ are found appear in Sec. II.B of Ref. [100].
TABLE V. $K_H^q$ values.

|       | $N$  | $\Delta$ | $\pi$  | $V$  | $\rho$ | $\omega$ |
|-------|------|----------|--------|------|--------|----------|
| Set 1 (used in Ref. [81]) | 0.064<sup>a</sup> | 0.041<sup>a</sup> | 0.498<sup>a</sup> | 0.03 | 0.021<sup>b</sup> | 0.034<sup>b</sup> |
| Set 2 (more recent) | 0.048<sup>c</sup> | 0.020<sup>d</sup> | 0.494<sup>c</sup> | 0.058<sup>c</sup> |

<sup>a</sup> From Ref. [102], Eq. (85).
<sup>b</sup> From Ref. [103], Table 2.
<sup>c</sup> From Ref. [98], Sec. 2.
<sup>d</sup> From the RL2 with pion exchange result for $\sigma_{\pi\Delta}$ in Eq. (16) of Ref. [104].

Dimensionless response coefficients

$$K^H_E = \frac{m_H}{E} \frac{\delta E}{\delta m_H},$$

where $E$ is the unperturbed ground state energy [in Refs. [81] and [100], $K^H_E$ is denoted by $\Delta E(m_H)$]. The changes in $m_H$ are related to changes in $m_q$ by the sigma terms

$$\sigma_H = m_q \frac{d m_H}{d m_q} \equiv K^q_H m_H$$

inferred from studies of hadronic structure. Thus, in terms of the ground state energies $E_g = \langle g|\hat{H}|g \rangle$ and $E_{\tilde{g}} = \langle \tilde{g}|\hat{H}|\tilde{g} \rangle$,

$$\tilde{\sigma}_g \equiv m_q \frac{dS}{dm_q} = K_{E_g} E_g - K_{E_{\tilde{g}}} E_{\tilde{g}}, \quad (10)$$

where, via the chain rule for rates of change, the sensitivity

$$K_E = \sum_H K^H_E K^q_H,$$

the sum being over the hadrons identified above.

Relevant values of $K^H_E$ and $K^q_H$ are given in Tables IV (upper half) and V, respectively. Barring $K^q_\pi$, recent values of the coefficients $K^q_H$ (Set 2) are appreciably different from those used in Ref. [81] (Set 1). There is also the issue of what to adopt for $K^q_V$. The value chosen in Ref. [81] is the average (to one significant figure) of $K^q_\rho$ and $K^q_\omega$ in Set 1. We can make a similar estimate of $K^q_V$ with the value of $K^q_\rho$ in Set 2; we appeal to the fact that, in each of the two calculations [102, 103] known to us in which both $K^q_\rho$ and $K^q_\omega$ are determined, $K^q_\omega - K^q_\rho = 0.013$: consequently, our preferred value of $K^q_V$ is

$$K^q_V = \frac{1}{2}(2K^q_\rho + 0.013) = 0.06$$
to one significant figure. In view of the uncertainty in $K_V^q$, we have generated three sets of values of $K_E$ (sets 2a, 2b and 2c in Table IV) using set 2 of $K_H^q$’s in Table V and $K_V^q = 0.6$, 0.7 and 0.8, respectively. For comparison, the sensitivities quoted in Ref. [81] are included as set 1 in Table IV.

The corresponding values of $\tilde{\sigma}_s$ are presented in Table VI. The choice of Hamiltonian is such that ground state energies coincide with binding energies and we have used binding energies taken from experiment in evaluating Eq. (10). In view of the scatter of values in Table VI, no firm conclusions about the order of magnitude of $\tilde{\sigma}_s$ are possible, except that the estimate in Ref. [81] of $|\tilde{\sigma}_s| \sim 10$ MeV is an overestimate. In the case of set 2a in Table VI (our preferred choice), $|\tilde{\sigma}_s| \lesssim 1$ MeV.

Unfortunately, as inspection of the $K_E^V$ coefficients in Table IV and the manner in which changes in $m_V$ are effected (cf. Sec. II.B in Ref. [100]) reveals, $\tilde{\sigma}_s$ is most sensitive to that part of the AV18+UIX Hamiltonian which is least well connected to the properties of specific hadrons. However, there exists a framework for systematically relating phenomenologically successful nuclear forces to effective field theories appropriate to low-energy QCD [105–107]. A characterization of the short range part of the AV18 potential in terms of the low-energy constants (LECs) of an effective Lagrangian is known [108], and, recently, the quark mass dependence of LECs for the $^1S_0$ and $^3S_1 - ^3D_1$ partial waves has been established [98]. It is, perhaps, not too much to hope that some fruitful combination of these developments may circumvent the issues thrown up by the vector meson $V$. There remains, of course, the treatment of the three-nucleon UIX potential but contributions to the $K_H^q$’s from its

| TABLE VI. $\tilde{\sigma}_S$ in Eq. (10) for some $A = 5$ to $A = 9$ nuclei (using experimental binding energies $E_g^{\exp}$). |
| --- |
| $E_g^{\exp}$ | $^5$He | $^6$He | $^6$Li | $^7$He | $^7$Li | $^7$Be | $^8$Be | $^9$Be |
| --- | --- | --- | --- | --- | --- | --- | --- | --- |
| $\tilde{\sigma}_S$ (set 1)$^a$ | -3.42 | -9.92 | -9.52 | -11.7 | -15.4 | -15.5 | -17.2 | -16.2 |
| (set 2a) | -0.75 | -0.60 | -1.39 | -2.01 | 0.23 | -0.62 | 1.67 | -3.94 |
| (set 2b) | 0.17 | 2.57 | 1.45 | 1.06 | 5.20 | 3.83 | 7.77 | 0.17 |
| (set 2c) | 1.09 | 5.73 | 4.29 | 4.12 | 10.2 | 8.29 | 13.9 | 4.29 |

$^a$ From Table IV in Ref. [81] ($\tilde{\sigma}_s = m_q \delta S_{\exp} / \delta m_q$ in the notation of Ref. [81]).
two-pion part have been found to be small [100].

The Flambaum-Wiringa conjecture is an important idea, arguably a *sine qua non* for the reliable extraction of information on $X_q$ from Oklo data. More evidence in support of this conjecture is essential. Microscopic calculations for medium-heavy nuclei with the renormalized Fermi hypernetted chain method [109] would be a challenging but helpful line of investigation.

### C. Unified treatment of the sensitivities to $\alpha$ and $X_q$

On the basis of the results in the two previous subsections, we postulate the following relation for the shift $\Delta_r \equiv E_r(\text{Oklo}) - E_r(\text{now})$ in the position of a resonance (near threshold) due to (small) changes $\Delta X_q \equiv X_q(\text{Oklo}) - X_q(\text{now})$ and $\Delta \alpha \equiv \alpha(\text{Oklo}) - \alpha(\text{now})$ in $X_q$ and $\alpha$, respectively:

$$\Delta_r = a \frac{\Delta X_q}{X_q} + b \frac{Z^2}{A^{4/3}} \frac{\Delta \alpha}{\alpha},$$

(11)

where the coefficients $a$ and $b$ are independent of $A$ and $Z$. The lack of any dependence on $A$ and $Z$ in the first term is conditional on the validity of the Flambaum-Wiringa conjecture. The scaling with $A$ and $Z$ of the second term is, in part, deduced from Eq. (8) on substitution of the mass number dependence of $[r^2]_{gg}$ implied by the uniform shift formula (cf. Eq. (48) in Ref. [110]). We also have to assume that the integral discarded in Eq. (7) to obtain Eq. (8) shares this scaling.

Our deliberations in subsection VI A suggest it is reasonable to assume that $|b| \sim 1$ MeV. The order of magnitude of $a$ is less certain: according to Ref. [81], $a \sim 10$ MeV, but the results in Table VI indicate that $a$ could be one or even two orders of magnitude smaller ($a$ is the average of $-\bar{\sigma}_S$ for a given set of $K^0_N$'s).

With a large enough data set, the different dependences on mass and proton number in Eq. (11) should permit one to disentangle the contributions of $\Delta X_q$ and $\Delta \alpha$ without any assumptions about their relative size. Independent limits on $\Delta X_q$ and $\Delta \alpha$ would open up the possibility of constraining the mechanism for time variation along the lines pursued in Refs. [111], [112] and [113]. Apart from Ref. [71] (which invokes a questionable “mean scaling” hypothesis), we do not know of any Oklo studies which involve more than a couple of nuclei.

An entirely model independent limit on either $\Delta X_q$ or $\Delta \alpha$ is not possible for a *single*
nucleus, unless one of the two terms in Eq. (11) is known a priori to be dominant. In the case of $^{149}$Sm, it has been customary to discard the $X_q$-term, a step which has been strongly criticized on two grounds. First, there are studies (notably, Ref. [81]) which find that the coefficient $a$ of the $X_q$-term is an order of magnitude larger than the coefficient multiplying $\Delta \alpha/\alpha$; second, calculations [91, 92] based on the Callen-Symanzik renormalization group equation suggest that, within any theory which admits the unification of the Standard Model gauge couplings (at a mass scale $\Lambda_u$ below the new physics responsible for the time variation of fundamental constants), the evolution of these couplings to lower energies is such that $|\Delta_X/X_q|$ is an order of magnitude larger than $|\Delta \alpha/\alpha|$ if the behavior of other Standard Model parameters is ignored (and $\Lambda_u$ is not time-dependent). These objections do not stand up to closer inspection.

Our results on $a$ and $b$ indicate that the coefficients of $\Delta X_q/X_q$ and $\Delta \alpha/\alpha$ in Eq. (11) are more likely to be comparable. As regards the relative magnitudes of $\Delta X_q/X_q$ and $\Delta \alpha/\alpha$, the most complete statement implied by the analysis of Ref. [91] is (cf. Eq. (30) in Ref. [91])

$$\left| \frac{\Delta X_q}{X_q} \right| \sim (R - \lambda - 0.8\kappa) \frac{\Delta \alpha}{\alpha},$$

(12)

where $R \simeq \pi/[9\alpha(M_Z)]$, $\alpha(M_Z)$ being the electromagnetic coupling at the electroweak scale $M_Z$ [$\alpha(M_Z)^{-1} = 127.9$], and $\lambda$ and $\kappa$ parametrize the time-dependence of Yukawa couplings to fermions and the vacuum expectation value of the Higgs boson, respectively.\footnote{Cf. Eqs. (11) and (14), respectively, in Ref. [91]. More precisely, $\lambda$ denotes the average value of the $\lambda_u$'s in Eq. (11) of Ref. [91].} Equation (12) should be compared with the result in Ref. [91] for the variation in $\mu \equiv m_p/m_e$, namely

$$\frac{\Delta \mu}{\mu} \sim (R - \lambda - 0.8\kappa) \frac{\Delta \alpha}{\alpha},$$

(13)

[cf. Eq. (20) in Ref. [91], where $Y$ is used instead of the more standard notation $\mu$], and the experimental results

$$\frac{\Delta \alpha}{\alpha} = (-7.4 \pm 1.7) \times 10^{-6}$$

(14)

and

$$\frac{\Delta \mu}{\mu} = (2.6 \pm 3.0) \times 10^{-6}$$

(15)

for overlapping red shifts $1.8 < z < 4.2$ and $z \sim 2.81$, respectively [87, 114]. If the expression for $\Delta \mu/\mu$ obtained on combining Eqs. (13) and (14) is to be compatible with the bound in
Eq. (15), then the value of the (constant) factor $R - \lambda - 0.8\kappa$ must be such that $|\Delta X_q/X_q| \sim |\Delta\alpha/\alpha|$. Thus, under circumstances in which the rather general model of Ref. [91] applies, it is permissible to ignore the $\Delta X_q/X_q$-term in Eq. (11) when extracting an order of magnitude limit on $\Delta\alpha/\alpha$, and *vice versa*.

### D. Bound on the energy shift $\Delta_r$ of the Sm resonance

The experimental basis for extracting a bound on the energy shift is illustrated in Fig. 4 which shows the overlap of a simple Breit Wigner resonance with a thermal neutron spectrum. If the energy of the Sm resonance shifts down from its present day value of $E_r(\text{now}) = 97.3\text{meV}$, then more $^{149}\text{Sm}$ will be burned in the neutron flux, and less $^{149}\text{Sm}$ will be found in the isotopic remains of the reactor. Conversely, if the resonance shifts up, less will be burned, and the remains will be richer in $^{149}\text{Sm}$. The result is parametrized

![Diagram](image)

**FIG. 4.** Overlap of the 97.3 meV resonance of $^{149}\text{Sm}$ with a thermal neutron spectrum. If the energy of the resonance is different now compared to 2 Gyr ago, burn up of $^{149}\text{Sm}$ is changed and the isotopic abundances remaining today will in principle indicate the magnitude and sign of the energy shift.
by an effective capture cross section $\hat{\sigma}$ which will depend on the amount the resonance is shifted.

In practice, the temperature of the neutron spectrum must be taken into account, other resonances may have to be included, and details of the results can change when the epithermal component of the flux is included. This is illustrated in Figs. 5 and 6 which show $\hat{\sigma}$ values from Refs. [73] and [41], respectively. The former are calculated with thermal fluxes of different temperatures, the latter using realistic fluxes. While the shapes are similar, the differences in magnitudes are relevant when it comes to comparing with experimentally derived $\hat{\sigma}$ values.

The cross sections are obtained by solving coupled equations [41, 73] which seek to reproduce the isotopic abundancies reported for samples from the reactor zones [26]. The equations take into account isotope production in fission, generation of plutonium through neutron capture, and isotope burn up in the neutron flux. Post processing contamination is also sometimes included as an additional parameter in analyses [73].

As is to be expected from geological samples, while trends of isotopic depletion and change are clear, variations outside of the statistical uncertainties are seen and further complicate the extraction of a resonance shift. In Ref. [41] this was taken into account for RZ10 by not analyzing each sample individually but, instead, analyzing a meta sample, the average of the isotopic data for the four samples.

FIG. 5. Effective cross sections for $^{149}$Sm as a function of the 97.3 meV resonance shift, calculated with thermal neutron spectra of various temperatures. From Y. Fujii et al., Nucl. Phys. B573 (2000) 377, with permission from Elsevier.
FIG. 6. Effective cross sections for $^{149}$Sm as a function of the 97.3 meV resonance shift, calculated with neutron spectra that include both thermal and epithermal components. The shapes are similar to those derived from thermal only calculations (see earlier), but differ in magnitudes. From C. R. Gould, E. I. Sharapov and S. K. Lamoreaux, Phys. Rev. C 74 (2006) 24607.

The effective RZ10 neutron capture cross section for $^{149}$Sm determined in this way was $(85.0 \pm 6.8)\text{kb}$ and, as seen in Fig. 7, this leads to two solutions for the energy shift $\Delta_r$: a right branch overlapping zero,

$$-11.6\text{ meV} \leq \Delta_r \leq 26.0\text{ meV},$$

and a left branch yielding a non-zero solution, $-101.9\text{ meV} \leq \Delta_r \leq -79.6\text{ meV}$.

Similar double-valued solutions were found in Ref. [73], where gadolinium isotopic data were used to try and establish one or other of the solutions as more probable. An implication of Eq. (11) is that $\Delta_r$ should be the same to within a percent of so for the Sm and Gd data. Without the benefit of Eq. (11), the authors of Ref. [73] had to make the seemingly ad hoc assumption that Sm and Gd should give the same result for $\Delta_r$. The analysis of Ref. [73] favored the zero shift solution but was complicated by the post processing contamination issues mentioned earlier. At this time, while nearly all analyses are consistent with a zero shift, the non-zero solution of the left-hand branch is not ruled out on experimental grounds.
FIG. 7. Effective cross sections for $^{149}$Sm at 200 °C and 300 °C, and bounds (vertical and horizontal lines) indicating allowed solutions for the energy shift based on isotopic abundance data. From C. R. Gould, E. I. Sharapov and S. K. Lamoreaux, Phys. Rev. C 74 (2006) 24607.

Perhaps, Eq. (11) can be the basis for a strategy to decide conclusively on the interpretation of the left-hand branch of solutions.

E. Limit on $\Delta \alpha$ and $\Delta X_q$ implied by bound on $\Delta r$ for the Sm resonance

The data on root-mean-square charge radii in Table XII of Ref. [115] implies that the isotopes $^{149}$Sm and $^{150}$Sm have equivalent charge radii of 6.4786(10) fm and 6.5039(12) fm in their ground states $|g\rangle$ and $|\tilde{g}\rangle$, respectively. (The more recent but less precise data in Table 6 of Ref. [116] are compatible with these results.) As the $^{150}$Sm compound nucleus state $|r\rangle$ is just above the neutron escape threshold, the excitation energy is about 0.4 MeV per valence nucleon. This fact, in conjunction with the subshell spacing (in the vicinity of the Fermi levels) of the single particle level schemes for $^{150}$Sm (see, for example, Figs. 4 and 5 in Ref. [117]), leads us to conclude that the charge distribution of $|r\rangle$ will not be
significantly different from that of the ground state. Certainly, we do not anticipate a 25% increase in the equivalent charge radius to 8.11 fm (the value adopted in Ref. [72]). Instead, we set \( R_r = 6.5 \text{ fm} \), i.e. the value of the equivalent charge radius for the \(^{150}\text{Sm}\) ground state. According to the samarium data in Table X of the compilation in Ref. [115] (which supersedes the data of Ref. [93] used by Dyson and Damour),

\[
[r^2]_{gg} = 0.250 \pm 0.020 \text{ fm}^2.
\]

Equation (8) then evaluates to (with \( Z = 62 \))

\[
\left| \frac{dE_r}{d\alpha} \right| \sim 2.5 \text{ MeV}.
\]

Despite the substantially smaller choice of \( R_r \) (which is the smallest physically acceptable one), this revised estimate of \( |\alpha dE_r/d\alpha| \) is still of the same order of magnitude as the lower bound on \( \alpha dE_r/d\alpha \) used in Ref. [72].

Taking into account that Eq. (8) is an overestimate [because of the omission of the cancellation discussed in connection with Eq. (7)], we advocate the use of the relation

\[
\left| \frac{dE_r}{d\alpha} \right| \sim 1 \text{ MeV}
\]  

(17)

in the analysis of the \(^{149}\text{Sm}\) data. In effect, Eq. (17) differs from the standard result of Ref. [72] only in that there is no longer any attempt to attach a confidence level.

Equation (17), together with the bound on \( \Delta_r \) in Eq. (16) and the relation \( \Delta_r \simeq (\alpha dE_r/d\alpha)(\Delta\alpha/\alpha) \), implies the bound

\[
\left| \frac{\Delta\alpha}{\alpha} \right| < 3 \times 10^{-8}.
\]  

(18)

If the coefficient \( a \) in Eq. (11) is of the order of 1 MeV (the case for set 2a of sensitivities \( K_E \) in Table IV), then a similar bound applies to \( \Delta X_q/X_q \).

Since the publication of Ref. [72], it has been common practice to use the relation

\[
\frac{\Delta\alpha}{\alpha} = -\frac{\Delta r}{M}
\]  

(19)

with \( M = 1.1 \text{ MeV} \) to infer a bound on \( \Delta\alpha \) from a bound on \( \Delta r \). Section III.C of Ref. [40] can be consulted for a comprehensive overview of results based on Eq. (19) up to the publication of Ref. [41]. Table VII below contains some features of this summary and updates it to include Refs. [41] and [42]. It should be clear from our reappraisal of Ref. [72] (in subsection
VI A) that we believe one should be a little circumspect about the way Eq. (19) has been used in the past to restrict $\Delta \alpha$ and the average value of $\dot{\alpha}$ to intervals about zero. Order of magnitude estimates based on Eq. (19) are, however, probably reliable. Thus, we advocate reporting the result of, for example, Ref. [41] as the bound in Eq. (18). This bound is reduced by a factor of 3 in Ref. [42].

Our guarded attitude towards Eq. (19) is shared by the authors of Ref. [118]. In assessing the limits on $\dot{\alpha}/\alpha$ of Refs. [40] and [41] (in the last column of Table VII), they adopt the most conservative null bound of $|\dot{\alpha}/\alpha| \leq 3 \times 10^{-17} \text{yr}^{-1}$ and argue that, by multiplying this bound by a factor of 3, they can compensate for the neglected effect of variations in $X_q$ (and any other parameters influencing nuclear forces). This factor of 3 is arbitrary (as pointed out in Ref. [118]), but its use is taken for granted in subsequent studies [119]. We do not understand the stated rationale for the factor of 3, but it can be viewed as an \textit{ad hoc} way of accommodating partial cancellations between the $X_q$ and $\alpha$ contributions to $\Delta_r$.

| Ref. | Zones | Neutron spectrum | $\Delta \alpha/\alpha (10^{-8})$ | $\dot{\alpha}/\alpha (10^{-17} \text{yr}^{-1})$ |
|------|-------|------------------|---------------------------------|---------------------------------------------|
| [72] | 2,5   | Maxwell          | $-9 \rightarrow 11$            | $-5.5 \rightarrow 4.5$                   |
|      |       |                  | (180 – 700°C)                  |                                            |
| [74] | 10,13 | Maxwell          | $-2 \rightarrow 0.2$          | $-0.1 \rightarrow 1$                     |
|      |       |                  | (200 – 400°C)                  |                                            |
| [40] | 2     | MCNP4C$^b$       | $-5.6 \rightarrow 6.6$        | $-3.3 \rightarrow 2.8$                   |
|      |       |                  | (Fresh core)                   |                                            |
| [41] | 2,10  | MCNP4C           | $-1.1 \rightarrow 2.4$        | $-1.2 \rightarrow 0.6$                   |
|      |       |                  | (Spectral indices)$^c$         |                                            |
| [42]$^d$ | 3,5   | MCNP4C           | $-1.0 \rightarrow 0.7$        | $-0.4 \rightarrow 0.5$                   |
|      |       |                  | (Realistic fuel burn-up)       |                                            |

* Limits on the \textit{average} rate of change of $\alpha$ over the time since the Oklo reactors ceased (relative to the current value of $\alpha$). We take the age of the natural reactors to be 2 billion years.

* Spectrum of neutrons calculated with the code documented in Ref. [39].

* Model for spectrum consistent with measured Oklo epithermal spectral indices.

* The inequalities in Eq. (9) of Ref. [42] need to be reversed.
VII. CONCLUSIONS

Unravelling how the geosphere and the biosphere evolved together is one of the most fascinating tasks for modern science. The Oklo natural nuclear reactors, basically formed by cyanobacteria two billion years ago, are yet another example of the surprises to be found in Earth’s history. Since their discovery over forty years ago, the reactors have provided a rich source of information on topics as applied as can nuclear wastes be safely stored indefinitely to topics as esoteric as are the forces of physics changing as the Universe ages?

In this review, we have summarized nuclear physics interests in the Oklo phenomenon, focusing particularly on developments over the past two decades. Modeling the reactors has become increasingly sophisticated, employing Monte Carlo simulations with realistic geometries and materials which can generate both the thermal and epithermal fractions. The water content and the temperatures of the reactors have been uncertain parameters. We have discussed recent work pointing to lower temperatures than earlier assumed. Nuclear cross sections are input to all Oklo modeling and we have identified a parameter, relating to the capture by the $^{175}$Lu ground state of thermal neutrons, that warrants further investigation.

The use of Oklo data to constrain changes in fundamental constants over the last 2 billion years has motivated much recent work. We have presented a critical reappraisal of the current situation, starting with the long-standing study of Damour and Dyson on sensitivity to the fine structure constant $\alpha$. We conclude that their result can plausibly be used for order of magnitude estimates, but an investigation of how this conclusion may be affected by a more careful treatment of the Coulomb potential in the vicinity of the nuclear surface (and beyond) is warranted. The more recent analysis by Flambaum and Wiringa of the sensitivity to the average mass $m_q$ of the light quarks has been updated to incorporate the latest values of sigma terms. No firm conclusions about the reliability of Flambaum and Wiringa’s estimate are possible (because of uncertainties surrounding the short-ranged part of the nuclear interaction), but it could be an overestimate by as much as an order of magnitude.

On the basis of the work in Refs. [72] and [81], we have suggested a formula for the unified treatment of sensitivities to $\alpha$ and $m_q$, namely Eq. (11). It is an obvious synthesis, which has the advantage of making explicit the dependence on mass number and atomic number. We hope that it may prove useful in distinguishing between the contributions of $\alpha$ and $m_q$.
in a model independent way or, at least, facilitating an understanding of the significance of the non-zero shifts in resonance energies which have been found in some studies of Oklo data.

Appealing to recent data on variations in $\alpha$ and the proton-to-electron mass ratio $\mu$, we have demonstrated that, within the very general model of Ref. [91] (and contrary to widespread opinion), shifts in resonance energies are not any more sensitive to variations in $m_q$ than they are to variations in $\alpha$. When extracting an order of magnitude limit on any change in $\alpha$, it is, thus, permissible to ignore any changes in $m_q$ and vice versa (provided the model of Ref. [91] applies). In fact, we have argued that one can, at best, use null shifts to establish order of magnitude estimates of upper bounds. Bounds on $\Delta \alpha/\alpha$ have been presented. The most recent study [42] of the Oklo data pertaining to the 97.3 meV resonance seen now in neutron capture by $^{149}$Sm implies that $|\Delta \alpha/\alpha| \lesssim 1 \times 10^{-8}$ (cf. Table VII for more details).

ACKNOWLEDGEMENTS

We would like to thank V. V. Flambaum and R. B. Wiringa for responding to our queries about Ref. [81], and R. B. Wiringa for recalculating some of the entries needed in Table IV. C. R. G. acknowledges support by the US Department of Energy, Office of Nuclear Physics, under Grant No. DE-FG02-97ER41041 (NC State University).

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