Search for time modulations in the decay constant of ^{40}K and ^{226}Ra at the underground Gran Sasso Laboratory

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A B S T R A C T

Time modulations at per mil level have been reported to take place in the decay constant of several nuclei with period of one year (most cases) but also of about one month or one day. On the other hand, experiments with similar or better sensitivity have been unable to detect any modulation. In this letter we give the results of the activity study of two different sources: ^{40}K and ^{226}Ra. The two gamma spectrometry experiments have been performed underground at the Gran Sasso Laboratory, this way suppressing the time dependent cosmic ray background. Briefly, our measurements reached the sensitivity of 3.4 and 3.5 parts over 10^6 for ^{40}K and ^{226}Ra, respectively (1 sigma) and they do not show any statistically significant evidence of time dependence in the decay constant. We also give the results of the activity measurement at the time of the two strong X-class solar flares which took place in September 2017. Our data do not show any unexpected time dependence in the decay rate of ^{40}K in correspondence with the two flares. To the best of our knowledge, these are the most precise and accurate results on the stability of the decay constant as function of time.

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1. Introduction

A possible time dependence of the decay constant has already been discussed at the beginning of the science of radioactivity. As a matter of fact, at the end of the Ph.D. thesis of Marie Curie one finds the description of the search, with negative results, for a difference in the activity of uranium ores between midday and midnight. Recently, in particular since 2009 [1], various experiments have reported evidence of a time modulation of the decay constant of several radioactive nuclei, from ^{3}H to ^{239}Pu, with period, in most cases, of one year (but also of about one month or one day) and amplitude at the per mil level [2–4]. The annual modulation, with the maximum in February and the minimum in August, has been correlated to the change of the Sun–Earth distance between aphelion and perihelion. In [5,6] the interaction with solar neutrinos or the coupling to a long range scalar field from the Sun have been advocated as possible reason for the modulation. However, neutrino cross sections orders of magnitude higher than expected would be required. In addition, the laboratory constraints

on the variation of α_{em} and of the electron to proton mass ratio induce upper bounds to the relative variation of the decay constant nine orders of magnitude lower than the claimed per mil effect [7].

On the other hand, various experiments with similar or better sensitivity did not detect any modulation of the decay constants of several nuclei (a complete review is in [8]). Evidence against solar influences on nuclear decay constants in α, β^-, β^+ and electron capture decays [9–11] has been recently published using the data of 14 radionuclide metrology laboratories.

In the past we also performed a few gamma spectroscopy experiments in the underground Gran Sasso Laboratory, excluding modulations with amplitude larger than a few parts over 10^5 in ^{137}Cs [12], ^{222}Rn [13] and ^{232}Th [7].

2. The ^{40}K experiment

We already studied the decay constant of the electron capture decay of ^{40}K with an over-ground experiment. To our knowledge, this remains the only study on the subject. In particular, we measured a clear annual modulation with amplitude of $4.5 \pm 0.8 \cdot 10^{-5}$ and maximum at August 11th ($\pm$ 13 days). However, such an effect corresponds to a $\pm$ 3.5% modulation of the cosmic ray background (which we have measured) and it is well compatible, both in size
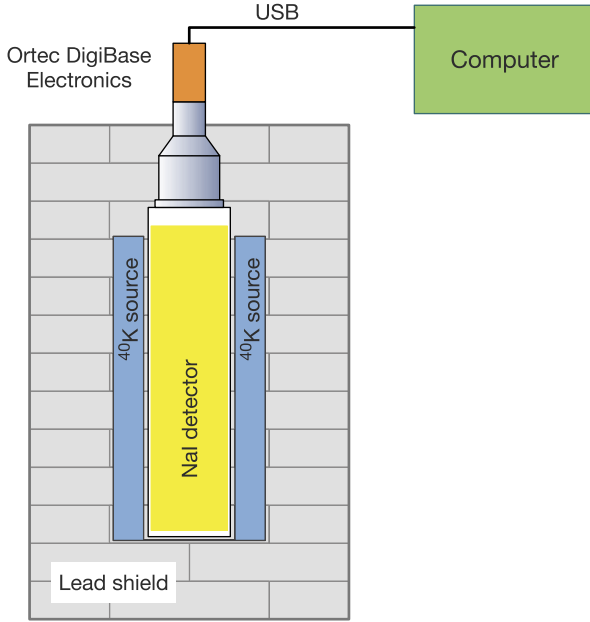
Fig. 1. The ^{40}K experimental set-up: the NaI crystal enclosed by the stainless steel box containing KHCO$_3$ melted with paraffine and shielded with 10 cm of lead.

Fig. 2. Measured γ-ray spectrum of the ^{40}K source and centroid position of the 1461 keV peak during the measurement (in the inset).

Fig. 3. Measured rate of the ^{40}K source averaged over 1 day. Horizontal lines are drawn at $\pm\,10^{-3}$ from the average. The shaded area corresponds to a 10^{-4} uncertainty.

and in phase, with the known annual modulation of the cosmic ray flux at the Earth surface (due to the change of the density in the upper atmosphere because of the temperature variation). In order to study the annual modulation of the ^{40}K decay independently of the comic ray flux, we built a new set-up which we installed deep underground in the Gran Sasso Laboratory, in a dedicated container of 2 m $\times$ 3.5 m and 2.5 m high placed in front of Hall B. As a matter of fact, the mountain shield suppresses the muon and neutron flux by six and three orders of magnitude, respectively, as compared to the above ground.

A 4 liter $4'' \times 4'' \times 16''$ NaI crystal detects the 1461 keV γ ray due to the electron capture decay of ^{40}K to the excited state of ^{40}Ar (10.55% branching ratio, half-life 1.25 $\cdot$ 10^9 y). The source is made of about 9.6 kg of potassium bicarbonate powder (KHCO$_3$, corresponding to 3.8 kg of natural potassium) and 2.4 kg of paraffine grains contained inside an hermetically sealed stainless steel box placed around the NaI detector. The bicarbonate and the paraffine grains have been well mixed together before being poured inside the stainless steel box. The box has then been kept inside an oven for about 20 hours at the temperature of 60 degrees Celsius. This way salt and paraffine melt together and, after cooling, a solid and compact block was formed. As a consequence it was possible to avoid the slow and long process of settlement of the salt powder (about 6 months) which was measured in [7], where KHCO$_3$ was used alone, and which was causing a time dependent detection efficiency. Finally, the whole set-up is shielded by at least 10 cm of lead (Fig. 1).

The electronic signals are processed by an Ortec (R) digiBASE (TM) with shaping time of 0.75 μs. In the analysis we consider the entire energy spectrum and not only the full energy peak at 1461 keV (Fig. 2). This because we want to avoid any inaccuracy coming from the fitting procedure and we also want to increase the total rate in order to improve the statistics. This procedure requires the definition of lower and upper boundaries. Since the content of the energy spectrum above ∼3 MeV is negligible, the only delicate point is the stability of the lower threshold that should be low enough to collect the entire spectrum and high enough to be well above the electronic noise. In addition, if the low energy threshold is sufficiently low and placed inside a flat region of the
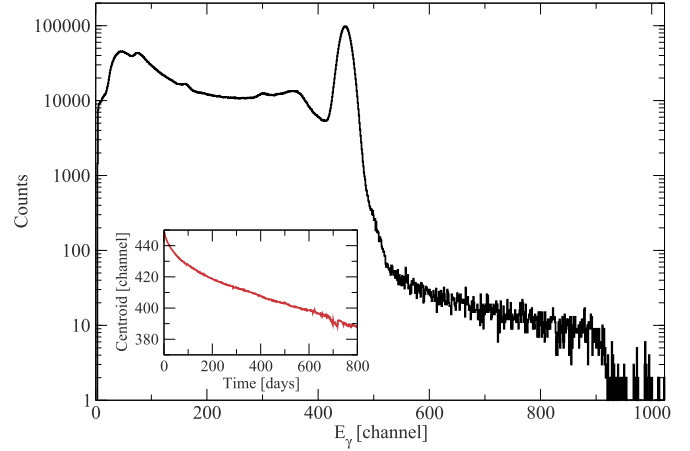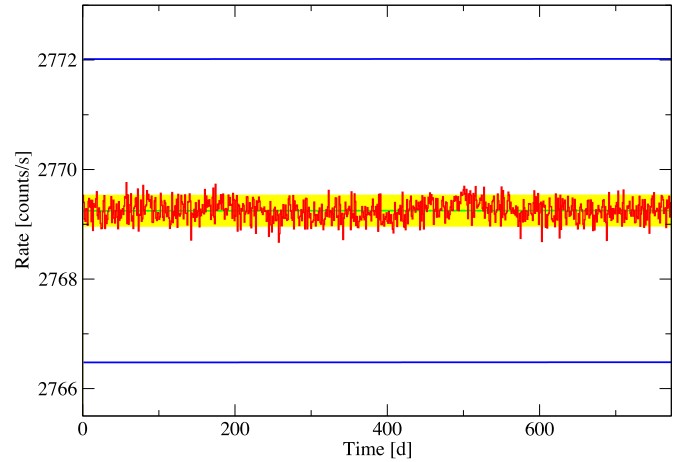
spectrum then it is also less sensitive to the variation of the global gain of the electronic chain. The intrinsic background, i.e. shielded set-up without the KHCO$_3$ salt, has been measured during a period of 12 days. Thanks to the underground environment and to the detector shielding, it is rather low, down to about 6.4 Hz above 15 keV.

Spectra are stored once per hour with a dead time of 1.24–1.25 $\cdot$ 10^{-2}, which has a fluctuation of 9 $\cdot$ 10^{-6} (1 sigma). The time when the system is unable to process input signals is essentially the ADC conversion time, i.e. the time required by the ADC to digitize the energy signals, and the time required by the electronics to store the event in the internal memory. This dead time is internally calculated by the digiBASE system and has been here normalized by the time that the spectrum was being recorded. The timing for data acquisition is provided by the internal quartz oscillator of the acquisition card. Its precision and stability (better than 10 ppm/year) are enough for our purposes. During the 799 days of data taking, from September 2015 to November 2017, we observed a monotonic change of the energy conversion gain (inset of Fig. 2): this produced a maximum shift of 61 channels for the ^{40}K peak at 1461 keV energy (initially at channel 449). The room temperature stayed within a 13.5–15 degree Celsius window.

The rate of about 2770 Hz is shown in Fig. 3 as function of time. We do not make any dead time correction to the data but we ap-