



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

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ABSTRACT

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 ^3H 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 (± 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

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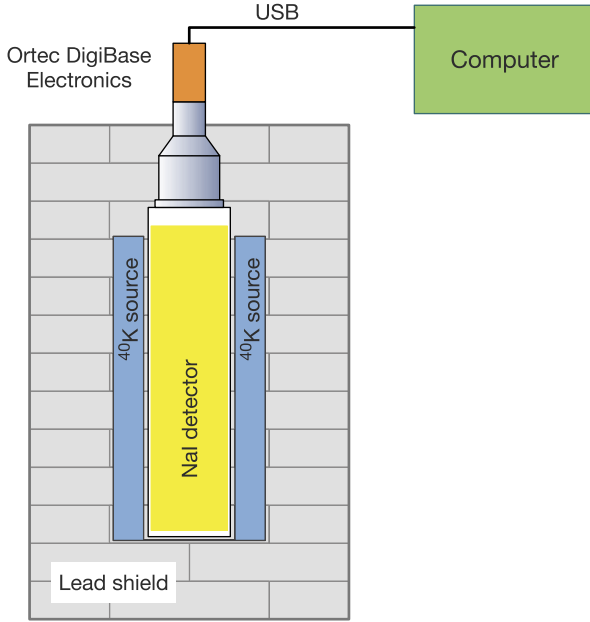


Fig. 1. The ^{40}K experimental set-up: the NaI crystal enclosed by the stainless steel box containing KHCO_3 melted with paraffine and shielded with 10 cm of lead.

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 cosmic ray flux, we built a new set-up which we installed deep underground in the Gran Sasso Laboratory, in a dedicated container of $2\text{ m} \times 3.5\text{ m}$ 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 $1.25 \cdot 10^9\text{ y}$). 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 $0.75\text{ }\mu\text{s}$. 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 $\sim 3\text{ MeV}$ 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

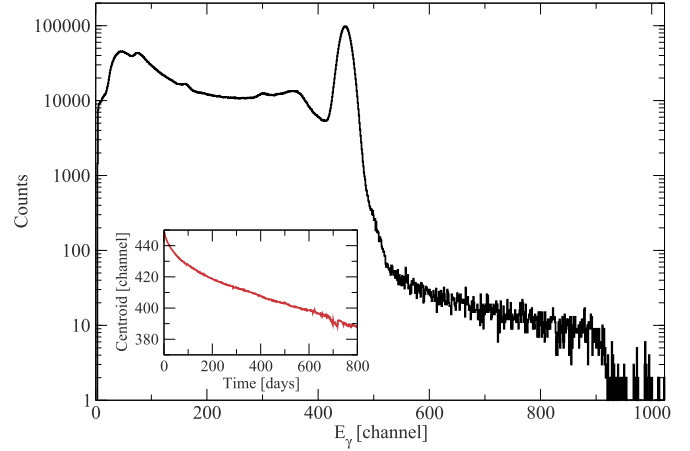


Fig. 2. Measured γ -ray spectrum of the ^{40}K source and centroid position of the 1461 keV peak during the measurement (in the inset).

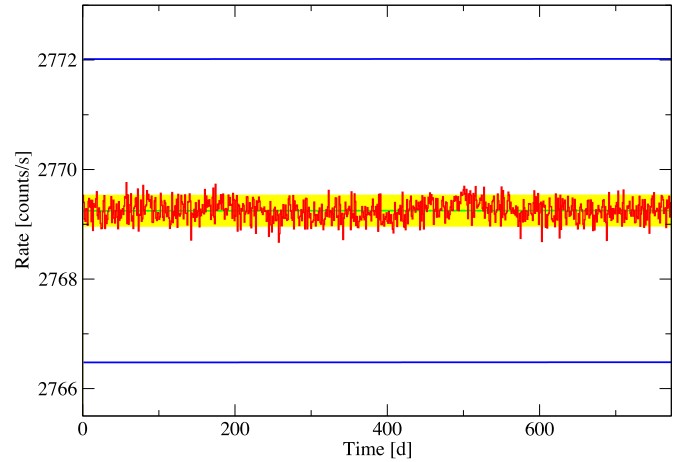


Fig. 3. Measured rate of the ^{40}K source averaged over 1 day. Horizontal lines are drawn at $\pm 10^{-3}$ from the average. The shaded area corresponds to a 10^{-4} uncertainty.

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 $1.24\text{--}1.25 \cdot 10^{-2}$, 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-

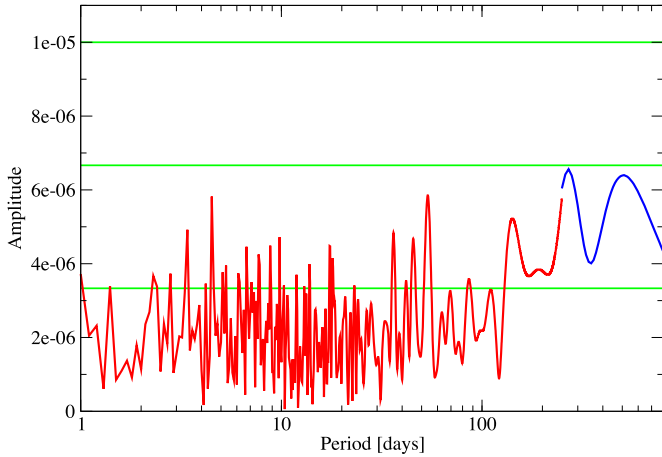


Fig. 4. Potassium source: amplitude of the different periods from the Fourier transform of the residuals and from the chi-squared analysis. Horizontal lines correspond to 1, 2 and 3 standard deviations, obtained with a 10000 time re-shuffling of the residuals or from the fit parameters.

ply a correction to the content of the first energy bin due to the peak position shift (at most 165000 counts/day, corresponding to 0.069% of the rate). The rate as function of time, when fitted with a constant and using only Poisson fluctuations as variance, gives a satisfactory chi squared per degree of freedom, χ^2/dof , of 1.11. At this point, the amplitudes of possible time modulations of the rate are searched for with the Fourier transform method (FFT) applied to the residuals or with the minimization of the chi squared fit of the residuals with a cosine function of time. As a matter of fact, Fourier transform can be rigorously applied only when searching for periods significantly smaller than the counting time. In particular, we have chosen a limit equal to one third of the run time. Finally, the statistical significance of the different amplitudes is obtained with a 10000 time re-shuffling of the residuals for the FFT analysis or directly from the errors on the fit parameters for the chi-squared analysis. Fig. 4 shows the results of the Fourier transform of the residuals, for periods from 1 to 250 days, and of the chi-squared analysis for longer periods.

In particular, using the chi-squared analysis of the residuals fitted with a cosine function of time we obtain the amplitude of $4.2 \pm 3.3 \cdot 10^{-6}$ (1 sigma) for the period of 1 year and phase unconstrained. The maximum amplitude provided by the chi-squared analysis is $6.6 \pm 3.3 \cdot 10^{-6}$ (1 sigma) for a period of 270 days and phase unconstrained. Both the amplitudes are not statistically significant.

Finally, we have studied the rate as function of the 24 hours of the solar and sidereal time (Fig. 5) to search for a possible daily modulation (i.e. 24 hour period and unknown phase). Both the rates are well compatible with a constant (χ^2/dof of 0.76 and 1.00, respectively) and time modulation amplitudes larger than $2.0 \cdot 10^{-6}$ and $2.3 \cdot 10^{-6}$ are excluded for the solar and sidereal time, respectively, by the chi-squared analysis of the rate with a constant fit.

3. The ^{226}Ra experiment

The set-up is installed in the same laboratory as the ^{40}K experiment. The source is a ^{226}Ra standard source (α decay + α/β decay of the chain) with Radium kept inside a sealed glass tube embedded in a plastic disk of 1" diameter and 1/8" thick. All the Radon produced from the decay of ^{226}Ra stays inside the glass tube, thus preserving the secular equilibrium. The source is kept

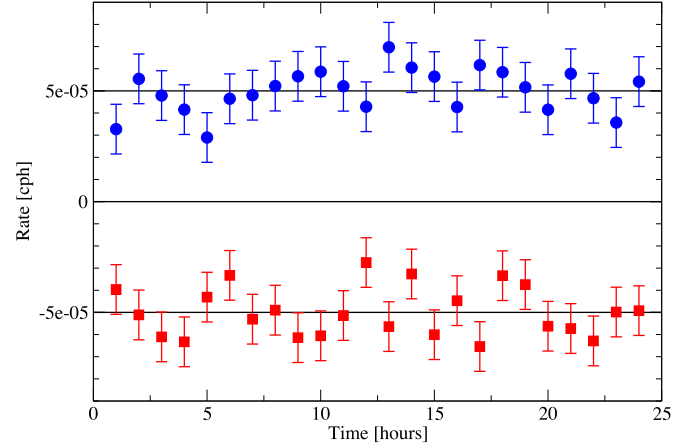


Fig. 5. Normalized residuals (averaged over the hour) of the ^{40}K source as function of solar (dots) and sidereal (squares) hours. Statistical uncertainties are indicated. An offset $\pm 5 \cdot 10^{-5}$ has been added to the datasets to improve the clarity.

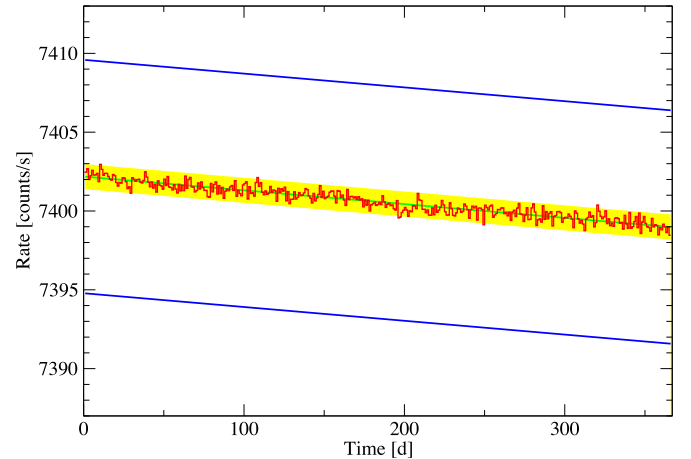


Fig. 6. Measured rate of the ^{226}Ra source averaged over 1 day. The two lines are drawn at $\pm 10^{-3}$ from the average. The shaded area corresponds to a 10^{-4} uncertainty.

at fixed distance from the flat face of the detector, a $3'' \times 3''$ NaI crystal horizontally placed.

Also in this experiment the signals are processed by an Ortec (R) digiBASE (TM) with shaping time of 0.75 μs and the entire energy spectrum is used to obtain the rate of the ^{226}Ra source, 7170 Hz above the 10 keV threshold. The intrinsic background has been measured during a period of 12 days. It amounts to 1.01 Hz above 10 keV.

Gamma spectra are stored every 3 hours with a dead time of $3.18 \cdot 10^{-2}$, which has a fluctuation of $4 \cdot 10^{-5}$. During the 1 year running time, from November 2015 to November 2016, the rate decreased by 0.032%, as expected from the 1600 y half-life of ^{226}Ra , the dead time decreased to $3.16 \cdot 10^{-2}$ and the peak at 1120 keV energy monotonically shifted by 39 channels. Because of this shift, we apply a correction to the content of the first energy bin (0.08% at most). The rate as function of time, corrected for the dead time, is shown in Fig. 6. When fitted with the expected decay law and using only Poisson fluctuations as variance, it gives a χ^2/dof of 1.32.

Fig. 7 shows the results of the Fourier transform of the residuals, for periods from 1 to 100 days, and of the chi-squared analysis for longer periods.

In particular, using the chi-squared analysis of the residuals fitted with a cosine function of time we obtain the amplitude of

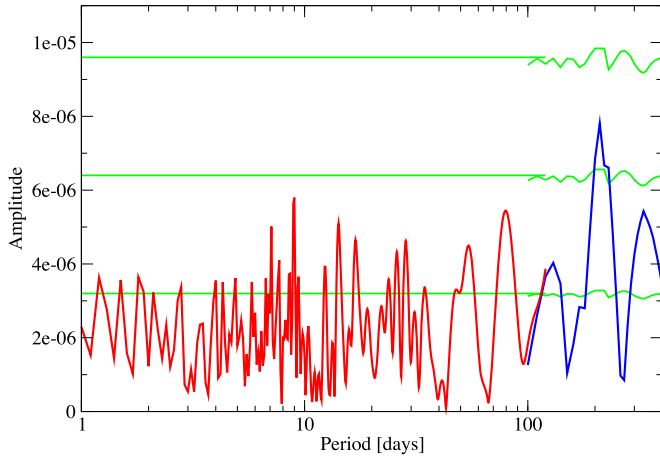


Fig. 7. Radium source: amplitude of the different periods from the Fourier transform of the residuals and from the chi-squared analysis. Horizontal lines correspond to 1, 2 and 3 standard deviations, obtained with a 10000 time re-shuffling of the residuals or from the fit parameters.

$4.7 \pm 3.2 \cdot 10^{-6}$ (1 sigma) for the period of 1 year and phase unconstrained. The maximum amplitude provided by the chi-squared analysis is $7.8 \pm 3.3 \cdot 10^{-6}$ (1 sigma) for a period of 210 days and phase unconstrained. Both the amplitudes are not statistically significant. We note that our sensitivity is a factor 300 smaller than the ^{226}Ra modulation discussed in [1] and one order of magnitude smaller than the best sensitivity quoted in [8].

4. Source activity during the two X-class solar flares of September 2017

Solar flares are explosions on the Sun that happen when energy stored in twisted magnetic fields (usually above sunspots) is suddenly released. This energy, up to one hundredth of the solar luminosity, is released within a few minutes to tens of minutes. Solar flares are classified according to the power of the X-ray flux peak near the Earth as measured by the GOES-15 geostationary satellite: X identifies the class of the most powerful ones, with a power at the peak larger than 10^{-4} W/m^2 (within the X-class there is then a linear scale).

In [6] a significant dip (up to $4 \cdot 10^{-3}$, $\sim 7 \sigma$ effect) in the count rate, averaged on a time interval of 4 hours, has been observed in the activity of a $\sim 1 \mu\text{Ci}$ source of ^{54}Mn (electron-capture) in coincidence with the X-3 and X-1 solar flares from December 2nd 2006 to January 1st 2007.

We already measured the decay constant during the occurrence of strong X-class solar flares in the years 2011–2013. No significant deviations from expectations have been observed larger than a few parts over 10^4 [7,14]. Here we give the results obtained by studying the activity of the ^{40}K source in correspondence with the two X-class solar flares which took place on the 6th and the 10th of September 2017 (X9.3 and X8.2, respectively, the most intense flares recorded during the current 11-year solar cycle, started on December 2008).

Fig. 8 shows the data collected in a 20 day window starting on the 25th of August 2017. In particular, the residuals of the count rate of the ^{40}K source are plotted, averaged over a period of 1 hour. The error bands are purely statistical since systematic uncertainties are negligible as compared to the statistical ones.

Our data clearly exclude an effect as large as the one reported in [6], of the order of a few per mil per day and lasting several days. In particular, the maximum effect compatible with our data is smaller than $6 \cdot 10^{-5}$ per day and $3 \cdot 10^{-4}$ per hour.

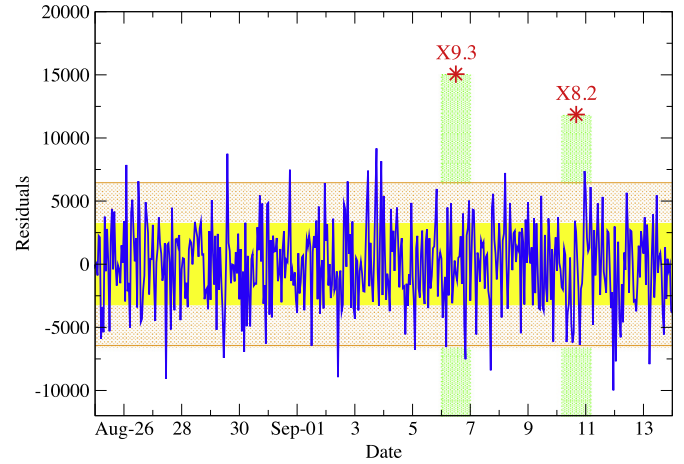


Fig. 8. Residuals, averaged over 1 hour, of ^{40}K data collected from the 25th of August 2017. Shaded areas correspond to 1 and 2 standard deviations from the expected value.

5. Conclusion

We have performed two gamma spectroscopy experiments in the underground Gran Sasso Laboratory to study the decay constant as function of time of ^{40}K (electron capture decay) and ^{226}Ra (α decay + α/β decay of the chain). Thanks to the deep underground location it has been possible to remove the time dependent cosmic ray background. Our measurements have a sensitivity of 3.4 (^{40}K) and 3.5 (^{226}Ra) parts over 10^6 and they do not show any evidence of time dependence statistically significant in the decay constant. We studied also the daily dependence of the ^{40}K decay constant on the solar and sidereal time: again no effect has been detected with a limit on the relative amplitude of $2.0 \cdot 10^{-6}$ (solar time) and $2.4 \cdot 10^{-6}$ (sidereal time). Finally, we exclude effects on the decay constant of ^{40}K of relative amplitude larger than $6 \cdot 10^{-5}$ per day and $3 \cdot 10^{-4}$ per hour during the two solar flares of September 2017, the most powerful flares of the current 11-year solar cycle.

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