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Precision long-term measurements of beta-decay-rate ratios in a controlled environment

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ABSTRACT

We report on measurements of relative beta-decay rates of Na-22, Cl-36, Co-60, Sr-90, Cs-137 monitored for more than one year. The radioactive samples are mounted in an automated sample changer that sequentially positions the five samples in turn, with high spatial precision, in front of each of four Geiger–Müller tubes. The sample wheel, detectors, and associated electronics are housed inside a sealed chamber held at constant absolute pressure, humidity, and temperature to isolate the experiment from environmental variations. The statistical uncertainty in the count rate approaches a few times 0.01% with two weeks of averaging. Other sources of error are on a similar scale. The data are analyzed in variety of ways, comparing count rates of the various samples on one or more detectors, and comparing count rates of a particular sample across multiple detectors. We observe no statistically significant variations in the ratios of decay rates, either annual or at higher-frequency, at a level above 0.01%.

1. Introduction

Several studies over the last decade have addressed the possibility of annual variations in nuclear decay rates. This was first noticed in detector calibration data from the Physicalisch Technische Bundesantalt (PTB) [1] and from Brookhaven National Laboratory (BNL) [2]. After systematic influences were accounted for, the radioactive decay rates showed regular variability at the few times 10^{-4} level (see Table 1 in Ref. [3]). This analysis inspired many follow-up studies, some of which observe annual and more rapid variations [4–14,3,15–20], and some of which do not [21–30]. It has been suggested that decay rates are influenced by both the proximity and activity of the Sun. Correlations with higher-frequency internal solar dynamics has also been identified [5].

Time-dependent variations in the decay rate, if existent, would likely require an explanation involving new physics outside the standard model. Variations would call into question the validity of the exponential decay of radioactive nuclei, potentially require modifications to radiation standards, and have important implications for geochronology and astrochronology [23]. There could also be important applications. If the variations are related to the Solar neutrino flux, for example, it might be possible to use the variations as a neutrino detector, or perhaps to measure or predict solar flares [6,20].

Recent studies that discount the likelihood of a solar influence on decay rates have offered the following arguments: 1) Seasonal environmental variations can influence the performance of radiation detectors [31,22]. Depending on the detector type, these variations can be as large as 0.1% [32]. Decay measurements in Am and Eu, for example, show that the seasonal variations in these two elements are highly correlated, but detector-specific. Some detectors show more seasonal variation than others [23]. This indicates the importance of understanding and controlling detector errors, something that is especially important at the sub-percent level. 2) A survey of 67 decay-rate data sets, covering 24 isotopes decaying by alpha, beta-minus, beta-plus, or electron capture shows that most isotopes have at least one data set for which the seasonal variation in the activity rate is less than 0.01% [23]. The remaining 43 data sets showed variations above this level. The discrepancies in the data sets are consistent with argument 1) above.

In this paper, we present an analysis of newly measured decay-rate ratios. The apparatus was described recently in Ref. [32]. The setup is designed specifically to remove seasonal influences in detector sensitivity by tightly controlling absolute pressure and temperature of the detector environment. Moreover, the sample-changing system allows us to divide out remaining detector-based biases by taking ratios of count-rate measurements. From our measurements of five radioactive samples, we construct 10 unique

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decay-rate ratios. We are unable to detect statistically significant oscillations in the decay rate ratios at any frequency with a period of a year or less above the 0.01% level. This data adds to a growing body of evidence suggesting that solar-related variations in the nuclear activity must be below a fractional level of 0.01%.

2. Experimental description

In our experiment, the radioactive samples, detectors, and electronics are housed in a sealed chamber. The pressure inside the chamber is controlled to be 93.33 kPa, with an accuracy of 0.1 kPa, and the wall temperature of the chamber is controlled to be 32.2 °C with an accuracy of 1 °C. The gas inside the chamber is N_2, and the humidity ranges from 3% to 4%.

Five different samples are placed in bismuth-lined sample holders mounted in an aluminum wheel. The wheel sequentially rotates each sample into position above four Geiger–Müller tube (GM) detectors once each day. The sample position relative to the detector is regulated to within 0.01 mm. The samples are Na-22, Cl-36, Co-60, Sr-90, and Cs-137. These beta-emitters were chosen because they have shown different levels of variation in previously published studies. The sample wheel also contains an empty space so that the background signal level can be measured.

3. Geiger–Müller tube data

The four GM detectors sequentially measure beta emission from samples of Na-22, Co-60, Sr-90, Cs-137, and Cl-36. These isotopes have half-lives of 2.6029, 5.2711, 28.80, 30.05, and 302000 years, respectively [33]. These samples had an initial activity of nominally 1 μCi. We use plastic Delrin disks with different sized holes in front of the samples to limit the detector count rate for all samples to roughly 400 counts per second (cps). This count rate was chosen so that the statistical errors in the count rate would approach a level of 0.01% in two weeks of averaging.

Each sample is positioned over each detector for four hours each day. The count data are recorded at five-minute intervals. For every sample and detector combination, we compute the average count rate over the entire four hours, and then average 14 days together for one data point.

3.1. Deadtime correction

The deadtime of the GM tubes is in the neighborhood 0.2 ms, depending on tube operating parameters including internal pressure, voltage, detected radiation energy. This deadtime results in a deadtime correction to the count rates of about 8%. While this correction may seem high for measurements claiming to reach relative sensitivities at the 0.01% level, it is nominally the same for all of the samples on each detector. Therefore in the count rate ratio, the relative importance of the deadtime correction is reduced.

Different mathematical models can be used to estimate the number of counts arriving during the avalanche recovery [34]. In our data, we correct the measured count rate R_m by assuming a deadtime τ in order to find the estimated "true" count rate R_t using the formula

$$R_t = \frac{R_m}{1 - R_m \tau}. \tag{1}$$

As mentioned, because the count rates are similar for all of our samples, the particular details of the model are comparatively less important. We first estimate the deadtime τ using the traditional additive method. In our data analysis, we also make small corrections in the deadtimes τ so that our fitted decay rates match the known values [33]. The resulting deadtimes on our four detectors are determined to be 0.250, 0.185, 0.259, and 0.181 ms. These values are consistent with those we measured using the additive method.

3.2. Ratio measurements reduce systematic variability

The measured count rate for each sample-detector combination depends on the source activity, detector sensitivity and gain, discriminator levels, and quantum efficiency. The count rate is also influenced by geometric and environmental factors, such as the proximity of the detector to the sample, sensitivity to ambient pressure, electrical charging of the sample disk, and so forth. In our previous work, we showed that typical seasonal variations in ambient pressure, for example, change the GM tube count rate by typically 0.15% per kPa, depending on both the detector and the energy of the detected beta particle. Dark signals, background levels, and deadtime also influence the measured count rate.

A significant advantage of our experiment is that it allows us to compute count rate ratios rather than being restricted to individual sample-detector data. In these ratios, nearly all of the factors mentioned above divide out or are significantly minimized. This is illustrated in Fig. 1. This plot shows relative count rate data for both Na-22 and Cl-36 as a function of time (upper left plot) after deadtime correction and background subtraction. Fitting the data using the known exponential decay rate reveals a slow time-dependent variation in the residuals (lower left plot). The data in this plot are averaged across all four detectors. The residuals for the individual detectors vary significantly, with trends that are somewhat steeper than the average shown in Fig. 1(b) to nearly flat. This suggests that trends in residuals are largely detector artifacts. However, when the ratio between different isotopes is taken, and the ratio data are fit to an exponential decay, this common drift in the residuals is absent, as seen in the two right-hand plots of Fig. 1. This analysis clearly shows the power of the ratio technique. Systematic detector-based variations are significantly reduced. This analysis is the same as that used in Ref. [2].

3.3. Ratio data analysis

Using five samples, we can calculate 10 unique count rate ratios for each detector. After deadtime correction and background subtraction, we calculate our count rate ratios and then fit each ratio to the exponential decay

$$s(t) = s_0 \exp(-\lambda t), \tag{2}$$

with both s_0 and λ as free parameters. Our results are shown in Table 1, compared to the ratios calculated from the known decay rates for each sample.

The decay rates in Table 1 are calculated using a linear, weighted least-squares fit to the log of the count rate ratio data. Each data point in the fit is the average of the ratio across the four detectors. The uncertainty in each data point for the fit is the standard deviation between the four detectors. The uncertainty indicated in the parentheses in column 3 of Table 1 is the 1σ estimated statistical uncertainty in the fitted decay constant [35].

For most of the data in Table 1, the 1σ statistical uncertainty in the fitted decay rate ratio is smaller than the difference between the known rate and our fitted rate. However, the differences follow a systematic trend. The data from Table 1 are plotted in Fig. 2. The vertical axis is the difference between our fitted decay rate and the known rates, $\lambda - \lambda_0$ from Table 1. The horizontal axis is the energy difference between the beta energy from the isotope in the numerator of the ratio minus the beta energy from the isotope in the denominator if the ratio, $\Delta E \equiv E_u - E_\ell$ from Table 1. The data

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