Search for Variations of $^{213}\text{Po}$ Half-Life

E.N. Alexeyev, Yu.M. Gavrilyuk, A.M. Gangapshev, A.M. Gezhaev, V.V. Kazalov, and V.V. Kuzminov
Baksan Neutrino Observatory INR RAS, Neitrino 361609, Russia
S.I. Panasenko and S.S. Ratkevich
V.N. Karazin Kharkiv National University, Kharkiv 61022, Ukraine

A device with the parent $^{229}\text{Th}$ source was constructed to search for variations of the daughter $^{213}\text{Po}$ half-life ($T_{1/2} = 4.2 \mu s$). A solar-daily variation with amplitude $A_{S0} = (5.3 \pm 1.1) \times 10^{-4}$, a lunar-daily variation with amplitude $A_L = (4.8 \pm 2.1) \times 10^{-4}$, and a sidereal-daily variation with amplitude $A_S = (4.2 \pm 1.7) \times 10^{-4}$ were found upon proceeding the data series over a 622-day interval (from July 2015 to March 2017). The $^{213}\text{Po}$ half-life mean value is found to be $T_{1/2} = 3.705 \pm 0.001 \mu s$. The obtained half-life is in good agreement with some of the literature values obtained with great accuracy.

Keywords: half-life, $^{213}\text{Po}$ nucleus, daily and annual variations

I. INTRODUCTION

Experimental studies of $^{214}\text{Po}$ half-life time dependence ($\tau$) \cite{1} have been carried out at the Baksan Neutrino Observatory of the Institute for Nuclear Research of the Russian Academy of Sciences since 2008. Unlike studies on determining the half-life by the results of analysis of time dependence of the explored isotope activity, decay curves analyzed at the Observatory are plotted based on a set of data on lifetimes of separate nuclei of the $^{214}\text{Po}$ isotope. In order to determine this parameter, the delays between the moment of a nucleus production (a beta electron from $^{214}\text{Bi}$ decay + a gamma-quantum) and its decay (an alpha particle from the $^{214}\text{Po}$ decay) are measured. The measurements are performed at TAU-2, a low-background facility placed in the underground low-background laboratory NLGZ-4900 at the depth of 4900 m.w.e. (973 days) and at TAU-1 of the underground low-background KAPRIZ laboratory at the depth of 1000 m.w.e. (354 days). The time interval of measurements at TAU-1 corresponds to the end of the measuring interval at TAU-2. Further, time series of $\tau$ with different temporal steps are analyzed. According to data obtained at TAU-2, the averaged value of the $^{214}\text{Po}$ half-life is $\tau = 163.47 \pm 0.03 \mu s$. The annual variation with amplitude $A = (9.8 \pm 0.6) \times 10^{-4}$, the solar-daily variation with amplitude $A_{S0} = (7.5 \pm 1.2) \times 10^{-4}$, the lunar-daily variation with amplitude $A_L = (6.9 \pm 2.0) \times 10^{-4}$, and the sidereal-daily variation with amplitude $A_S = (7.2 \pm 1.2) \times 10^{-4}$ are detected in the series of $\tau$ values.

It was found that $\tau$-amplitude maxima are observed at the moments when the maximum projection is reached by the Earths surface point velocity vector directed at the explored source of possible variations (the Sun, the Moon, or an unidentified stellar object). Basically, it would be possible to explain the origin of solar and lunar variations by these objects influence on performance of measuring facilities through cyclical geophysical and climatic disturbances (tidal waves, meteorological factors, magnetic field, etc.) caused by them on the Earth. However, no reasonable fundamental process capable of transforming these disturbances into variations of time parameters of measuring facilities in the required phase has been yet discovered.

At the same time, the sidereal-daily variation of $\tau$ observed, if this is not an instrumental effect, may identify the presence of a real unknown physical phenomenon influencing the parameter under study. Two checks have been performed to verify the reliability of the results. The dependencies of the amplitude and phase of the observed sidereal-daily wave on the choice of starting point of the analyzed time series were verified. The start was shifted by 91 days and 182 days. The previous series section was not considered in the analysis. As expected for this type of variation, waves were obtained with amplitudes coinciding with the initial values within the error, but with phases shifted by 6 and 12 hours relative to the initial phases. The second verification was performed by plotting the daily data set in an anti-sidereal time (nonexistent periodicity). The day duration in the anti-sidereal time was increased relative to solar day duration by an interval shortening the day duration in sidereal time. In such a data set, within the statistical error $\pm1.2 \times 10^{-4}$, the wave was absent.

Measurements were carried out at the TAU-1 facility to confirm the non-random character of the observed variations of the $\tau$ temporal series. Data provided by TAU-1 revealed a solar-daily variation with amplitude $A_{S0} = (17 \pm 3) \times 10^{-4}$, a lunar-daily variation with amplitude $A_L = (8 \pm 3) \times 10^{-4}$, and a sidereal-daily variation with amplitude $A_S = (11 \pm 4) \times 10^{-4}$. Searching for annual variations, initial data for a half year were summed and then sequentially shifted to increase the statistical reliability of the decay curve. As a result, a $\tau$ series of no more than six-month duration was obtained from the annual data set. Hence, it was impossible to identify the annual periodicity with adequate accuracy, so it was not explored. It is clear that a substantial improvement in the statistical error can be achieved by a substantial increase in the data-taking rate. However, in the case of...
the $^{214}$Po isotope, the speedup of data-taking induces a quadratic increase in the share of random coincidences, up to $\sim 1\%$ at a rate of $12\ s^{-1}$. This is caused by a high aggregate activity of all daughter $^{220}$Ra isotopes and the relatively long half-life of $^{214}$Po ($\sim 163.5\ \mu s$). Therefore, without augmentation of the relative contribution of the random coincidence background, the increase in the statistics accumulation rate for $^{214}$Po can be achieved only by the increase in the number of independent setups. This might appear technologically impossible. Another alternative is to use a pair of radioactive isotopes having a similar decay scheme, but essentially shorter half-life of the daughter isotope. The following isotopes might be used as such pairs: $^{213}$Bi ($T_{1/2} = 46\ \text{min}$) $\rightarrow$ $^{213}$Po (weighted average of $T_{1/2} = 3.72(2)\ \mu s$) that are daughter products in the series of decays $^{226}$Ra ($T_{1/2} = 7340\ \text{years}$) from the series of $^{237}$Np decays $^{[4]}$. In this paper, the first results obtained from using the facility with the specified source are presented.

II. THE FACILITY DESCRIPTION

The construction of the TAU-3 facility with a $^{229}$Th source is similar to TAU-1 and TAU-2 $^{[1]}$. It comprises a scintillation detector D1, a plastic scintillator (PS), which is made of two disks $d = 18\ \text{mm}$ and $h = 1\ \text{mm}$ glued together. The radiation source $^{229}$Th ($T_{1/2} = 7340\ \text{years}$) positioned between the disks is the parent isotope for $^{213}$Po. The test sample is manufactured at the Khlopin Radium Institute (St. Petersburg). The source is precipitated from $\text{Th(NO}_3)_4$ salt solution on the surface of a LAVSAN film with $h = 2.5\ \mu m$ and covered by the same film pasted along the edge by the epoxy resin. The assembly is placed at the bottom of a case made of VM-2000 reflecting film open from one end. The case is put inside a stainless steel rectangular case $9 \times 23 \times 140\ \text{mm}$, thickness $0.5\ \text{mm}$. The open end of the case is connected with the bottom of a 2.5-mm stainless steel cylinder with $d = 44\ \text{mm}$, and $h = 160\ \text{mm}$. Inside the cylinder, there is a high-speed FEU-87 photomultiplier monitoring PS. The signal is taken from the FEU anode load through the matching circuit and supplied via the cable (50 Ohm) to the first entry of the registering detector. Detector D1 is placed in the 15-cm Pb protective layer in a gap with $h = 10\ \text{mm}$ between two scintillation detectors NaI(Tl) $150 \times 150\ \text{mm}$ (detector D2) in a low-background box of the NLGZ-4900 underground low-background laboratory $^{[6]}$. Signals from the anodes of two photomultipliers of the D2 detector are amplified by charge-sensitive preamplifiers, summed, and supplied to the second, starting entry of the registering detector. The registering facility comprises a LA-n10-12 PCI digital oscilloscope (DO) integrated with a PC that is registering in online mode the waveform of pulses arriving from D1 and D2. The frequency of pulse digitization in DO is chosen as 100 MHz. The reading and recording is started by a pulse in the D2 channel. The record frame is 2048 temporal channels (10 ns per channel), including 256 channels of prehistory and 1792 channels of history. In Fig. $^{[1]}$ the decays of $^{213}$Bi and $^{213}$Po isotopes $^{[8]}$ are presented schematically. From Fig. $^{[1]}$, it follows that $66\%$ of $\beta$ decays of $^{213}$Bi are transitions to the ground level, and $31\%$ to the excited level with an energy of $440\ \text{keV}$. The decay of this level is accompanied by a $\gamma$ quantum emission ($26\%$ per decay). The isotope of $^{213}$Po decays in $100\%$ of cases with emission of an $\alpha$ particle with an energy of $8537\ \text{keV}$. If the device registers all three particles released by the decay of the pair of isotopes, it is the event with three pulses. In this event, pulses coming from the $\gamma$ quantum and $\beta$ particle coincide instantaneously, and the pulse from the $\alpha$ is delayed. In Fig. $^{[2]}$ one of the events (frames) stored by DO in the PC memory is displayed as an example. The pulse on the upper beam (1) is a $\gamma$ quantum, the first pulse train on the lower beam (2) corresponds to a $\beta$ particle, and the second one to an $\alpha$ particle. The observed triple coincidences considerably reduce the contribution of background events accompanying decays of the remaining isotopes in the chain of decays of $^{229}$Th to the total counting rate of the facility. The activity of $^{229}$Th is $\sim 80\ \text{Bq}$. Alongside the main isotope there are small amounts of extraneous radioactive impurities in the

FIG. 1. Decay schemes of $^{213}$Bi and $^{213}$Po.
The rate of the event recording started by DO by D2 pulses with amplitudes of 380 keV was $\sim 27$ s$^{-1}$. The rate of recording useful events with parameters of all pulses corresponding to $^{213}$Po decay was $\sim 18$ s$^{-1}$.

Following from Fig. 2, signals from and particles are trains of short subpulses with total duration of up to $\sim 1$ µs, decreasing exponentially in frequency and amplitude. The trains can overlap at small delays between particles; therefore, the processing program should consider relations between the amplitudes of the first and subsequent subpulses in a train in order to unambiguously separate the delayed ($\beta \otimes \alpha$) coincidences.

The delays between pulses in channel D1 are determined by the results of processing the recorded oscillograms, and a decay curve of daughter isotope $^{213}$Po is plotted for the chosen time interval. The half-life determination is based on this curve. The sequential time series of this magnitude is plotted.

### III. MEASUREMENT RESULTS

Continuous measurements started at TAU-3 on July 9, 2015. The statistics for 622 days (March 2017) are processed. In Fig. 3, the decay curve of the $^{213}$Po isotope is given. The value of $\tau$ was obtained approximating the decay curve by function $F(t) = A \times \exp[-\ln(2)t/\tau] + b$ using the minimum $\chi^2$ test in the delay interval of 0.5-13.0 µs. It was found that $\tau = 3.705 \pm 0.001$ µs.

The primary data-consistent summation method was used to find possible periodic dependencies. This is the method of the interior moving average: to find harmonics in a data series an interval is chosen about 0.5 of the expected period, and the required parameter is determined for this interval; then the interval is shifted by one step and the procedure repeats.

In the studies of daily variations of the $^{213}$Po half-life depending on solar, sidereal, and lunar time, the length of the respective day was divided into 24 hours. The length of a sidereal and of a lunar day in the standard solar time is 23 hours 56 minutes 4.09 s and 24 hours 50 minutes 28.2 s, respectively. A period of 12 hours was chosen as an interval of averaging. The analysis of events was made as follows. We selected the events registered in the interval of 0-12 hours for the entire period study and determined the half-life values. After that, the interval was shifted by one hour and the procedure repeated. The results of the search of the daily variation in solar time are given in Fig. 4. Here, the result of approximation of the daily half-life dependence by the function $\tau(t) = \tau_0[1 + 3.4 \times 10^{-4} \sin(2\pi/24(t - 9))]$ (red curve) is displayed, where $\tau_0$ is the mean half-life; $\omega = 2\pi/24$ h$^{-1}$; $A = 3.4 \times 10^{-4}$ is the amplitude; $\phi = -3$ h is a phase shift of the initial point of the curve relative to 0 hours. The figure shows that the time dependence of the $^{213}$Po half-life is well described by a sinusoidal function. The period found is 24 hours and the relative amplitude is 0.00034 half-lives.
the time interval comparable to a year in the series of problem. The presence of a pulse surge of data within measurements can possibly provide the solution of this cast further curve tendency, and only further consistent instrumental effect, for example, equipment ageing, and a parameter are not clear yet. It could be both an in-

\[ \tau = \tau_0[1 + 4.2 \times 10^{-4} \sin\{2\pi/24(t - 1)\}] \] (blue dot-dashed curve).

It is easy to show that the initial periodic dependence of time data has the same period (24 h), the amplitude is higher by the factor of \( \pi/2 \) and is shifted by 0.5 of the moving interval (0.25 × 24 = 6 h). The amplitude of the initial daily periodic dependence obtained from these data in solar time is \( A_S = (5.3 \pm 1.1) \times 10^{-4} \) (blue dot-dashed curve).

In Fig. 5, the results of the search for a sidereal daily variation of the \(^{213}\)Po half-life are displayed. The experimental data are approximated by curve \( \tau(t) = \tau_0[1 + Asin\{\omega(t + \phi)\}] \) (red curve) with parameters \( A = 2.7 \times 10^{-4} \) is amplitude; \( \phi = -19 \) h is the phase shift of the curve initial point relative to 0 hours.

The analysis of the restored initial dependence similar to the analysis made for the solar-daily wave shows the presence of a sidereal-daily wave with relative amplitude \( A_S = (4.2 \pm 1.7) \times 10^{-4} \) (blue dot-dashed curve).

In Fig. 6, the results of search for a lunar-daily variation of the \(^{213}\)Po half-life are given. The analysis of the restored initial dependence like the analysis made for the solar-daily wave shows the presence of a lunar-daily wave with relative amplitude \( A_S = (4.8 \pm 2.1) \times 10^{-4} \) (blue dot-dashed curve).

In Fig. 7 the time dependence of \( \tau \) obtained from the decay curve for the weekly data set (start of measurements: July 9, 2015). weekly data hinders using the method of moving internal average for studies of the half-life annual variation. Therefore, in order to get a better understanding of the annual variation, we have checked a supposition that the half-life annual variation detected in the series of weekly data on the \(^{214}\)Po isotope continues with the same amplitude and phase in the series of data on \(^{213}\)Po. The data normalized to unity for these isotopes in continuous time are presented in Fig. 8. The data normalization for \(^{213}\)Po was made using the \( \tau \) mean value for 320 days. It is shown that the annual variation of the \(^{213}\)Po data with the same amplitude and phase as the data on \(^{214}\)Po is not excluded. The repeated deviation, after its shape is specified, can be removed from the series of data on \(^{213}\)Po to study the remainder for annual variations.
physical factors. To find the answer we need to continue measurements.

A component with frequency of 9.43 yr$^{-1}$ (period of 38.73 days) and the maximum power was detected over the period of June 1996–July 2001 in the analysis of the power spectrum of frequency components composing the series of counting rates ($\sim$ 1/5 day) at the Super-Kamiokande facility [7]. We have searched for a similar variation in our data series using the method of the interior moving average. The interval of averaging was chosen as 19.365 days with a one-day step. The result of processing is presented in Fig. 9 (triangles). The data were approximated by the function $\tau(t) = \tau_0[1 + A\sin\{\omega(t + \phi)\}]$, where $\tau_0$ is the mean half-life; $t$ is time [days]; $\omega = 2\pi/38.73$ day$^{-1}$; $A = (6.7 \pm 1.1) \times 10^{-4}$ is amplitude; $\phi = -17.4$ day is the phase shift of the curve initial point relative to zero. The restored wave amplitude $A = (10.6 \pm 1.9) \times 10^{-4}$ was obtained from the approximation by multiplying by $\pi/2$. In order to verify the result stability, a similar procedure was performed with the data of the TAU-2 facility accumulated for 500-day measurements with the $^{214}$Po isotope. The restored wave amplitude was $A = (10.6 \pm 1.9) \times 10^{-4}$. The search for a wave with the frequency of 10 yr$^{-1}$ was carried out for verification. Within the statistical error of $\pm 1.9 \times 10^{-4}$, no variation with this frequency was detected. Thus, we can conclude that the variation with the frequency of 9.43 yr$^{-1}$ is of global character (at least, for the underground devices), though does not coincide with known natural rhythms. In [7], the authors consider the possibility that the Sun’s core can feature a similar rhythm, and study possible mechanisms of these variations.

V. CONCLUSIONS

In this study, to search for variations of $^{213}$Po half-life ($T_{1/2} = 3.7$ µs), a device with $^{229}$Th isotope as a parent source was constructed. A solar-daily variation with amplitude $A_S S = (5.3 \pm 1.1) \times 10^{-4}$, a lunar-daily variation with amplitude $A_L = (4.8 \pm 2.1) \times 10^{-4}$, and a sidereal-daily variation with amplitude $A_S = (4.2 \pm 1.7) \times 10^{-4}$ were discovered upon processing the data series for the period from July 2015 to March 2017 (622 days). The $^{213}$Po half-life value averaged over 662 days is found to be $T_{1/2} = 3.705 \pm 0.001$ µs. This is consistent with the result ($T_{1/2} = 3.708 \pm 0.008$ µs) obtained by means of an ion-implanted planar Si detector for alpha and beta particles emitted from weak $^{225}$Ac sources in work [8].

ACKNOWLEDGMENTS

The study was supported by the High Energy Physics and Neutrino Astrophysics Program of the Presidium of the Russian Academy of Sciences.
[1] E.N. Alexeyev, V.V. Alekseenko, Ju.M. Gavrjuk, A.M. Gangapshev, A.M. Gezhaev, V.V. Kazalov, V.V. Kuzminov, S.I. Panasenko, S.S. Ratkevich, and S.P. Yakimenko, “Experimental test of the time stability of the half-life of alpha-decay $^{214}$Po nuclei,” Astropart. Phys. 46, 23 (2013).

[2] E.N. Alexeyev, Yu.M. Gavriljuk, A.M. Gangapshev, V.V. Kazalov, V.V. Kuzminov, S.I. Panasenko, and S.S. Ratkevich, “Sources of the systematic errors in measurements of $^{214}$Po decay half-life time variations at the Baksan deep underground experiments”, Physics of Particles and Nuclei, 46, 157 (2015).

[3] E.N. Alexeyev, Yu.M. Gavriljuk, A.M. Gangapshev, V.V. Kazalov, V.V. Kuzminov, S.I. Panasenko, and S.S. Ratkevich, “Results of a search for daily and annual variations of the $^{214}$Po half-life at the two year observation period”, Physics of Particles and Nuclei, 47, 986 (2016).

[4] G. Audi, F.G. Kondev, M. Wang, B. Pfeiffer, X. Sun, J. Blachot, M. MacCornick, “The Nubase2012 evaluation of nuclear properties”, Chin.Phys. C 36, 1157 (2012).

[5] Reference book edited by I. K. Kikoin “Tables of physical magnitudes”, (Atomizdat, Moscow, 1976) [in Russian].

[6] Yu.M. Gavriljuk, A.M. Gangapshev, A.M. Gezhaev, V.V. Kazalov, V.V. Kuzminov, S.I. Panasenko, S.S. Ratkevich, A.A. Smolnikov, and S.P. Yakimenko, “Working characteristics of the New Low-Background Laboratory (DULB-4900)”, Nucl. Instr. Methods in Phys. Res. A 729, 576 (2013).

[7] P.A. Sturrock, E. Fischbach, and J.D. Scargle, “Comparative Analyses of Brookhaven National Laboratory Nuclear Decay Measurements and Super-Kamiokande Solar Neutrino Measurements: Neutrinos and Neutrino-Induced Beta-Decays as Probes of the Deep Solar Interior”, Solar Physics 291 3467 (2016).

[8] G. Suliman, S. Pommé, M. Marouli, R. Van Ammela, H. Stroh, V. Jobbágy, J. Paepen, A. Dirican, F. Bruchetseifer, C. Apostolidis, A. Morgenstern, “Half-lives of $^{225}$Fr, $^{217}$At, $^{213}$Bi, $^{213}$Po and $^{209}$Pb from the $^{225}$Ac decay series”, Applied Radiation and Isotopes 77 32 (2013).