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Precision half-life measurement of ^{140}La with Ge-detector

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Abstract

Half-life is one of the fundamental properties of radioactive nuclei, and the precision required for its numerous applications in modern physics sometimes approaches the level of 10^{-4}–10^{-5}. Most part of the $T_{1/2}$ measurements performed up to now was made with proportional chambers, and the results were sometimes hardly reproducible within the error limits. Using Ge-detectors for that purpose brought some significant advantages but electronic unit related effects and spectra analysis procedures still remain the sources of the errors influencing the accuracy of the $T_{1/2}$ attained.

In this work, ^{140}La samples were obtained in the ^{139}La$(n,\gamma)^{140}$La reaction, employing a microtron as a neutron source and the half-life measurements were performed with a HPGe-detector. Influencing factors such as photopeak and background shape, electronic circuitry dead time and deadtime variations during the measurements, as well as pulse pileup are studied altogether. Values of the ^{140}La $T_{1/2} = 1.6808(18)$ d, $\lambda = 0.47749(20) \times 10^{-5}$, agreeing within the uncertainities with the most accurate evaluated ones ($T_{1/2} = 1.6781(3)$ d, $\lambda = 0.47807(9) \times 10^{-5}$) [2] were obtained in two series of measurements. © 2002 Published by Elsevier Science B.V.

1. Introduction

The half-life together with the nuclear mass, Q-value, decay modes with its branching ratios are the basic decay characteristics of a given radioactive nucleus.

A high precision in half-life ($\sigma(T_{1/2})/T_{1/2} < 10^{-4}$ or better) is required for the following studies:

(a) Weak interaction. For example, superallowed $0^+ \rightarrow 0^+$ beta transitions between members of an isospin multiplet have the same ft value for

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all such transitions if the isospin is a good quantum number [1].

(b) The rate of electron capture is essentially proportional to the density at the nucleus of electrons available for capture in the nucleus. If we change this density by means of chemical composition we may observe differences in half-lives by about 10^{-4} or smaller.

(c) Neutron-rich La and Ba isotopes are well-known fission fragments from the $U(n, \text{fission})$ reaction. The isotop ^{140}La has intense high energy gamma rays, well suited for investigations with Ge-detectors. Its half-life of 40.272 h renders it well suitable for immediate industrial investigations of the amount of fissile material in spent-fuel.

(d) Requirements on the accuracy of the half-life determination are high when one needs to calculate the intensity of the radiation N_0 at the time $t(0)$ several half-life periods before the actual measurements of the radiation intensity $N(t)$ at later time (decay correction).

The aim of this work was to determine at best the half-live of ^{140}La with present-day standard γ-spectroscopic techniques using Ge-detectors and to find out what disturbing influences have to be considered in the final total error. The relative error $\sigma(N_0)/N_0$ is calculated from the expression for the radioactive decay

$$\sigma(N_0)/N_0 = \left\{ [\sigma(N(t))/N(t)]^2 + (t \ln 2/T_{1/2})^2 (\sigma(T_{1/2})/T_{1/2})^2 \right\}^{1/2},$$

assuming that the measurement error $\sigma(N(t))$ and the error $\sigma(T_{1/2})$ are independent. If one can neglect the relative measurement error $N(t)$, then $\sigma(N_0)/N_0$ is proportional to $(t \ln 2/T_{1/2})$, e.g., at $t = 5T_{1/2}$ one calculates $\sigma(N_0)/N_0 = 3.5(\sigma(T_{1/2})/T_{1/2})$. Thus, one may conclude that if it were possible to use standard γ-sources whose intensity is determined with an accuracy of $\pm 0.1\%$, one may determine the $T_{1/2}$ with an accuracy about 10^{-4}, when making the measurement over several half-life periods.

The half-life of ^{140}La was measured several times mainly with the help of the proportional chambers. A high precision in determination of the $T_{1/2}$ was obtained, and most measurements are in good agreement; however, sometimes results deviate by more than cited errors (see Table 1, based on [2–9]). The results of some earlier $T_{1/2}$ measurements for ^{140}La are given additionally in [7], but the accuracy of those measurements is about one order of magnitude lower than the data presented in Table 1. Most of the accurate data were obtained with ionization chambers and only in [5] the $T_{1/2}$ was measured by means of the Ge(Li) detectors. The value for $T_{1/2}$ given in [5] has the highest accuracy, however, the way how this accuracy was obtained was not discussed in detail.

Table 1
Data on $T_{1/2}$ definition for ^{140}La according to publications with the most accurate values

$T_{1/2}$ (d)	λ ($c^{-1} \times 10^5$)	Ref.
1.6781(3)	0.47807(9)	[2]
1.6783(7)	0.47802(20)	[3]
1.6779(4)	0.47813(11)	[4]
1.67850(20)	0.47796(6)	[5]
1.6783(3)	0.47802(9)	[6]
1.67900(20)	0.47782(6)	[7]
1.6780(3)	0.47810(9)	[8]
1.6789(5)	0.47784(14)	[9]

Comparing the related merits of gas-discharge 4π-detectors (low efficiency, insensitivity to electrons) with semiconductor detectors, the later show some significant advantages in half-lives measurement. First of all, it is the possibility of measuring $T_{1/2}$ over long times and control the changes of the area of specific gamma-lines during the measuring time. This solves to a large extent the problem of the source impurity activities and makes unnecessary some corrections, such as impurities, detector current time-stability, the necessity to compare with the calibration activity source, etc.

Additionally, we thought it is interesting to mention another reason to study this phenomenon: our team is also working on some problems associated with the study of ''Accelerator Driven Systems'' (ADS), as described in [10]. In this context we investigated the space distributions of the flux of thermal neutrons generated in interactions of relativistic protons with extended targets

using the following reaction as sensor for thermal neutrons:

$$^{139}\mathrm{La}(n,\gamma)^{140}\mathrm{La}\xrightarrow{\beta^-}{}^{140}\mathrm{Ce}. \quad (1)$$

This reaction was used to determine the flux of thermal neutrons from heavy Pb- and/or U-targets irradiated with the beams from the JINR Synchrophasotron of the Laboratory of High Energies (LHE) in particular with protons of energy 3.7 GeV ("LHE" – experiment) [10]. A total of 15 La samples were irradiated simultaneously. The half-life of ^{140}La was determined using well-known HPGe gamma detection techniques. We observed apparent differences in the half-lives for ^{140}La activities from these samples as compared to literature value. This literature value is [2]

$$T_{1/2} = 1.6781(3) \text{ days}$$
$$\text{or } \lambda = 0.47807(9) \times 10^{-5} \text{ s}^{-1}, \quad (2)$$

as compared to our measurement of

$$T_{1/2} = 1.7095(25) \text{ days}$$
$$(\text{or } \lambda = 0.46929(69) \times 10^{-5} \text{ s}^{-1}). \quad (3)$$

The difference between these two values is 12σ (standard deviations). This forced us to look carefully at all possible sources of the errors which could lead to such a discrepancy. Specific measurements were carried out to find the most precise value of the ^{140}La half-life attainable with our advanced analytical techniques and to investigate the causes of the discrepancy between the values mentioned above.

2. Method of measurement and half-life determination

Measurements were performed using the HPGe-detector (ORTEC) with relative efficiency of 28%, and an energy resolution 1.8 keV for the gamma line of 1332.5 keV (^{60}Co). The spectrometer was operated with a CANBERRA-2026 amplifier, the multi-channel analyzer (MCA) Spectrum Master 919 (ORTEC) having an analog-to-digital conversion time below 7 μs and the difference between "live" and total measurement time was taken care of. The MCA was supplied with an independent memory and was connected to the PC providing convenient operation control for all the circuits, visualization of the MCA operation and data storage in the PC. One can define the measuring time for "live" or "real" measurements.

A high accuracy of the data-handling requires a careful approach to the processing of the spectra. The method to define the gamma line, the proper background subtraction, and correctness of the error estimation will reflect in the accuracy and reliability of the result.

The half-life, $T_{1/2}$, is found from the decay constant λ using the expression

$$S_i\Delta t(i)_{\text{real}}/\Delta t(i)_{\text{live}} = (\mathrm{e}^{-\lambda t(i)} - \mathrm{e}^{-\lambda(t(i)+\Delta t(i)_{\text{real}})})N_0, \quad (4)$$

where S_i is the area of the peak in the ith measurement; $\Delta t(i)_{\text{real}}$ is the duration of the ith measurement (real measurement time); $\Delta t(i)_{\text{live}}$ is the live measurement time defined over the spectrometer readings; $t(i)$ is the absolute starting time of the ith measurement; N_0 is the starting activity of the source.

The correct measurement of the live time is one of the significant aspects influencing accuracy of employed method, where areas of separate spectral lines rather than the decrease in the general activity of the source is measured. We tested the correctness of the exposure "live time" of our Spectrum Master 919 spectrometer.

Relation (4) is correct under the assumption that the dead time during a measurement is constant and that one can neglect the counting losses due to pile-up. Assuming that the dead time of the installation is changing in proportion to the total activity in the sample, i.e.

$$t_{\text{real}} - t_{\text{live}} = a_1 - a_2\mathrm{e}^{-\lambda t},$$

and the probability of pile-up is proportional to the dead time (see details in [8]) one can take these corrections into consideration using the relation

$$\begin{aligned} S_i\Delta t(i)_{\text{real}}/\Delta t(i)_{\text{live}} = N_0\big\{&(\mathrm{e}^{-\lambda t(i)} - \mathrm{e}^{-\lambda(t(i)+\Delta t(i)_{\text{real}})}) \\ &+ C_1(\mathrm{e}^{-2\lambda t(i)} - \mathrm{e}^{-2\lambda(t(i)+\Delta t(i)_{\text{real}})}) \\ &+ C_2(\mathrm{e}^{-3\lambda t(i)} - \mathrm{e}^{-3\lambda(t(i)+\Delta t(i)_{\text{real}})})\big\}, \end{aligned} \quad (5)$$

where C_1, C_2 are the coefficients defined by the least squares method [9].

3. Experimental results

Two experiments to determine the half-life of ^{140}La were carried out using two sample holders for lanthanum chloride ($LaCl_3 \cdot 7H_2O$), each sample weighting about 1 g. They were exposed to the flux of thermal neutrons generated with a beam from the Microtron (Flerov Laboratory of Nuclear Reactions, JINR) [11]. We designated these experiments as MICRO1(M1) and MICRO2(M2).

The decay of ^{140}La activity was measured through the investigation of its seven most intense gamma lines at 328.8, 487.0, 751.6, 815.8, 867.8, 925.2 and 1596.2 keV. The half-life was evaluated for each line separately and the results were averaged with the proper statistical weights, corresponding to the errors of each measurement (see Table 2). We also calculated $T_{1/2}$ from the summing of the intensities of all seven lines in each spectrum as given in Table 3. Finally, we used integral counts of the gamma spectra registered in the region from 329 to 1598 keV and present the results in Table 4. This procedure could be applied because we have a high activity purity in the radioactive ^{140}La source.

Table 4
$T_{1/2}$ values for ^{140}La obtained by summing the number of spectral pulses in the energy range $328 < E_\gamma < 1599$ keV for the MICRO2 (M2) experiment as well as considering different numbers of exponential terms from Eq. (5)

M2-FIT	λ ($c^{-1} \times 10^5$)	χ^2
1-exp	0.4811(4)	1370
2-exp	0.47569(24)	105
3-exp	0.47765(17)	20

We compared the processing procedures using two different analytical approaches to define the net peak areas: one using DEIMOS [12] and the other by MAESTRO [13]. The DEIMOS program determines the shape of the gamma-line and the shape of the background line and finds the model parameters by the least squares method. The main fraction of the peak is described by a Gaussian. Deviations from the Gaussian on the left and on the right flank depend on the energy of the gamma-

Table 2
$T_{1/2}$ for the ^{140}La defined for each of the seven above mentioned spectral lines and averaged considering the weight of each result

	LHE		M1		M2		M1 + M2
FIT	λ ($c^{-1} \times 10^5$)	χ^2	λ ($c^{-1} \times 10^5$)	χ^2	λ ($c^{-1} \times 10^5$)	χ^2	λ ($c^{-1} \times 10^5$)
l-exp	0.4696(7)	0.9	0.47787(21)	5.6	0.47684(19)	4.2	0.4773(5)
2-exp	0.4709(11)	1.1	0.47739(26)	0.8	0.47754(20)	2.3	0.47749(16)
3-exp	0.4708(18)	2.0	0.4772(4)	1.3	0.47759(23)	2.1	0.47749(20)

The data for three measurements (LHE, M1, M2) and total M1 + M2 result are presented. Data dependence on the calculation using expression (5) considering one, two and three exponents – i.e. dependence of the result on the variations of the dead time during the exposition (2-exp) and additionally consideration of detector signals overlapping (3-exp) are shown.

Table 3
$T_{1/2}$ values for the ^{140}La for three experiments (LHE, M1, M2) obtained by summing the peak areas for each time point over all seven lines are given

	LHE		M1		M2	
FIT	λ ($c^{-1} \times 10^5$)	χ^2	λ ($c^{-1} \times 10^5$)	χ^2	λ ($c^{-1} \times 10^5$)	χ^2
l-exp	0.4705(17)	114.8	0.47812(24)	0.69	0.47664(11)	0.93
2-exp	0.4718(25)	116.3	0.4776(4)	0.64	0.47749(18)	0.27
3-exp	0.4730(33)	118.4	0.4776(6)	0.66	0.47767(21)	0.23

Dependence of the $T_{1/2}$ values on the variation of the dead time during the exposition and pulses overlapping: the second and the third exponents, respectively, are presented analogously to the Table 2.

quantum and on the counting rate of the Ge-detector. They were taken into consideration by inserting satellite peaks (additional Gaussians) in the shape of the line or by adding the tails described by different analytical functions, e.g. by an exponentially decaying tailing function.

This method (only one Gaussian) allows one, in the case of sufficient counting rates, to obtain a χ^2 value between 50 up to hundreds. In this case the error in determination of the area and the position of the peak is several times larger as compared to fit with additional Gaussians where χ^2 is on the order of unity. The shape of the background under the peak is calculated as the superposition of a step function and the linear and quadratic functions (DEIMOS).

The other method (program MASTER) automatically determines the peak position ($K_{\max}$), its half-width, a linear background averaged over three points from the left to the right at a distance of 2.5 half-widths and the area N_{peak} of the peak is the integral of the counts in the channels from $(K_{\max} - \text{FWHM})$ to $(K_{\max} + \text{FWHM})$ minus the background area. The error N_{peak} is calculated as the square root of N_{peak}. Evidently, this method determines the background less accurately and the error of the area definition is quite crudely underestimated.

However, different methods (DEIMOS and MAESTRO) provide the results which differ in the peak area up to 4% and the differences independent of the energy of the gamma-quanta. This leads to discrepancy beyond the error limits in the value of λ and correspondingly $T_{1/2}$.

However, at small counting rates after a long decay time this discrepancy between the two approaches practically disappears, the values coincide but give the $T_{1/2}$ significantly different from that for the points with larger statistical significance. Because of the low weight of the points with the "poor statistics" they practically do not influence the result of $T_{1/2}$. Significant difference between the $T_{1/2}$ values obtained from calculations of S_i using the DEIMOS or the MAESTRO programs indicate the importance of the accurate determination of the background under the peaks. We assume that the background subtraction method used in DEIMOS is more accurate.

4. Further experimental details

(1) The measurement conditions for the first microtron experiment (MICRO1) are as follows:
   (a) 30 min irradiation of the sample;
   (b) distance from the container to the end of the detector cryostat was 35 mm along the symmetry axis of the Ge-detector, and the sample container was not moved during the experiment;
   (c) a cobalt-60 source was placed at a distance to the detector along the symmetry axis of 150 mm. This source was also fixed during the entire measurement. The cobalt-60 source was used to control the accuracy of the assessment of the live time by means of the SPECTRUM MASTER 919 device because an error in its operation might be the reason of systematic inaccuracies and the discrepancy of our results from the reference value given in [2];
   (d) measurement time was 493.5 h (12.2 "$T_{1/2}$" periods);
   (e) dead time at the start was 15%, 5% of which was due to the rather strong cobalt-60 source. The measurements were performed over a live time of 40 000 s per point, thus yielding 40 spectra.

Conditions of the ^{140}La and ^{60}Co line processing are as follows: we used the manual version of the DEIMOS program. The ratio S_1/S_n of the areas of ^{60}Co is shown in Fig. 1, where S_1 is the first point, S_n is the subsequent point. The points indicate that one needs a correction for the live time or pile-up in the first three points because the ratio decreases and levels off later when the dead time is below 8%. We introduced corrections to the first three points accordingly (see Fig. 1) in order to get the final result.

The half-life of ^{140}La obtained from this first measurement (MICRO1) is

$$T_{1/2} = 1.6788(7) \text{ days}$$
$$\text{or } \lambda = 0.4772(4) \times 10^{-5}. \quad (6)$$

This measurement indicated that our analysis using the equipment, data processing and analysis as presented, gives a value for λ in agreement with the value for λ from [2] within two standard deviations.

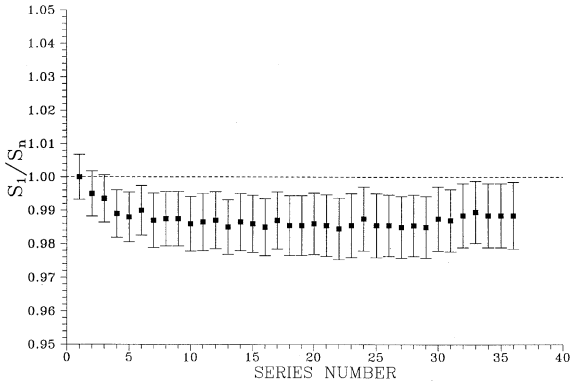

Fig. 1. Dependence of the relation of the S_1/S_n summarized areas of the two known ^{60}Co peaks on the number of the spectrum (measurement) accompanying by decreasing run rate (dead time). S_1 – the area of the peak in the first measurement, S_n – the peak area in the nth measurement.

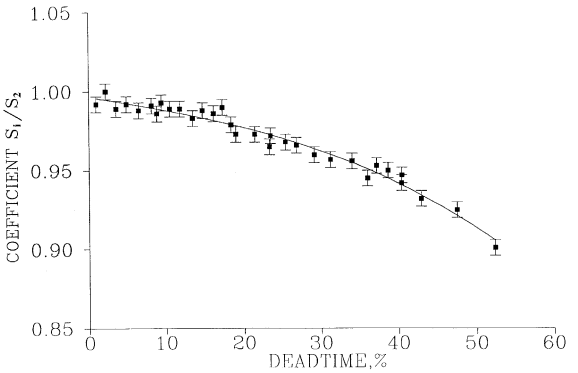

Fig. 2. Dependence of the S_i/S_2 relation on the total area of two peaks (1173, 1332.5 keV) ^{60}Co on the dead time. S_i – area of the peaks for the current point, S_2 – area of the peak corresponding to the minimal dead time.

(2) The measurement conditions for the second microtron experiment (MICRO2) are as follows:
   (a) all equipment is the same as in previous measurement;
   (b) for measurement the irradiated container is in close contact with the detector;
   (c) the dead time at the start is 53%. The measurement is carried out over the real time of 28 000 s for 42 spectra, consecutively 43 000 s for 11 spectra and finally 86 000 s for 2 spectra;
   (d) measurement time is 595 h or 14.8 half-life periods;
   (e) conditions of data processing and analysis are the same as in the previous case.

To study the operation of the MCA circuitry and to find possible corrections, we carried out methodological measurements. We measured spectra with two additional sources, ^{60}Co and ^{137}Cs. The cobalt source was fixed at a distance of 60 mm from the detector, the cesium source was moved with respect to the detector for each measurement in order to cover the entire range of dead time in the second measurement (MICRO2) i.e. from 53% to 0.95% (only the cobalt-60 source). The maximum load of the circuitry at a dead time 53% was 24 300 input pulses/s (counter) or 12 500 events/s in the spectrum. The minimum load (only ^{60}Co) was 405 input pulses/s or 277 events/s.

The ratio of the total area S_i of the ^{60}Co at 1173 and 1332 keV to their area S_2 at the minimum dead time as a function of the dead time is shown in Fig. 2. The behavior of the curve shows that the shift of the live time reaches 9% at the dead time of 52% (the measurements were carried out for a live time 2000 s). The curve from Fig. 2 was used to introduce corrections for the dead time in determining $T_{1/2}$ during this second measurement. The inaccuracies in the readings of the live- (or dead-)time from SPECTRUM MASTER 919 are determined with the CANBERRA 2026 amplifier in the spectrometric circuit. Analogous measurements with the CANBERRA 2020 amplifier showed the absence of such live time distortions: results analogous to those in Fig. 2 did not show the deviations of the ratio S_i/S_2 from unity. Thus, in order to estimate correctly the live- (dead-)times with the SPECTRUM MASTER 919 it is necessary to use an amplifier that provides the signals necessary for correct counting of the live time or to introduce additional live time control.

The over-all final results for all our measurements (LHE, MICRO1 and MICRO2) are given in Table 2. The MICRO2 measurement yields for the half-life of ^{140}La,

$$T_{1/2} = 1.68224(7) \text{ days}$$
$$\text{or } \lambda = 0.47759(23) \times 10^{-5} \text{ (Table 2; 3-EXP)}. \quad (7)$$

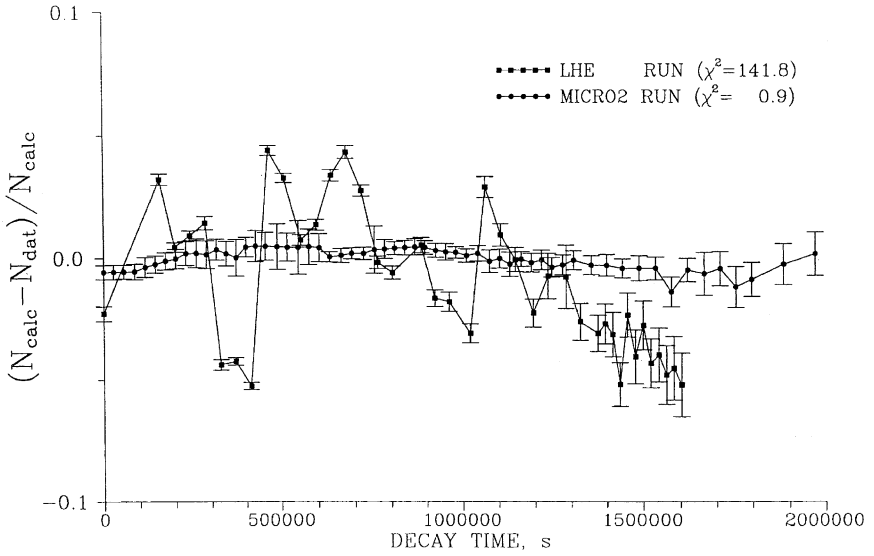

Fig. 3. Difference between the calculated values of the exponential behavior of the activity decrease and the experimental values in the measurement points for the LHE and MICRO2 experiments. The areas of the seven ^{140}La lines are summarized for each point.

The relative shift of the experimental values from the non-linear fit of the decay curve is shown in Fig. 3. It is seen that for the LHE experiment these values (filled squares) are characterized by a considerable discrepancy up to 4%,

$$[N_{\mathrm{calc}}(i) - N_{\mathrm{dat}}(i)]/N_{\mathrm{calc}}(i) = [\log S(i)_{\mathrm{calc}} - \log S(i)_{\mathrm{dat}}]/\log S(i)_{\mathrm{calc}},$$

which is appreciably larger than the statistical errors. The same data from the experiment MICRO2 are shown in Fig. 3 as black points for comparison. This measurement does not show the deviation from the non-linear fit. This discrepancy in the points can be explained with the non-uniformity of the powder in the container which can cause the effect due "changing efficiency of gamma-quanta registration" with the detector in close geometry of counting. (Remember: in the LHE experiment, the ^{140}La-container was taken out of the counting position after each counting period, to be replaced by another sample for counting in the extensive experiment as reported in [10]. During the specific MICRO experiments, the sample container remained unchanged in the same position during the entire counting period.) The observed deviations shown in Fig. 3 for the LHE-irradiation can thus be understood.

The final result of the two determinations of the decay constant for ^{140}La using neutrons from the Microtron is

$$T_{1/2} = 1.6808(18) \text{ days}$$
$$\text{or } \lambda = 0.47749(20) \times 10^{-5} \quad (8)$$

(see Table 2, M1 + M2, 3-EXP).

5. Conclusions

The following essential results have been presented in this publication:

(1) By measuring the half-life of nuclei with Gedetectors one can obtain the same high accuracy as has been obtained in measurements using proportional chambers. For this purpose one should
   (a) use an adequate method for background subtraction under the peaks: one must consider besides the linear and quadratic in the analytical functions also the "step function background",

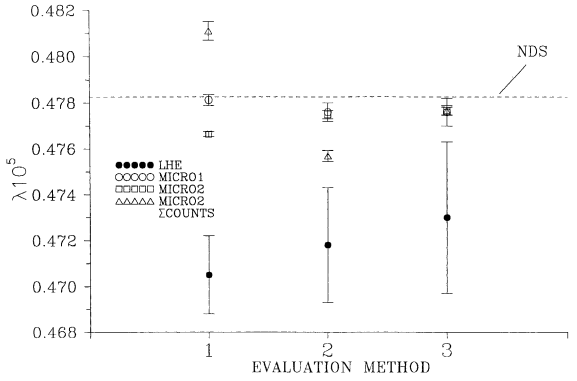

Fig. 4. Results of λ determination for the ^{140}La decay for all three measurements (LHE, MICRO1 and MICRO2) using different exponential terms from Eq. (5). The numbers 1–3 on abscissa determine the number of exponential terms of the expression (5) used for the λ calculation. The dashed line shows the value of λ from [2].

(b) make corrections for the changes of the dead time during the measurements and for the influence of the pile-up in the γ-line intensity (see Fig. 4). If we use Eq. (5) with its second and third exponential terms we come close to the reference result of [2] to within two standard deviations. However, there are big discrepancies if we use only the first term.

(2) One must take into account slight "variations in the geometric registration efficiency" of the Ge-detector in our measurements with the $LaCl_3{\cdot}7H_2O$ powder during the sample changing ("LHE experiment"). A possibly uneven activity distribution in the sample may lead to systematic errors in determination of the half-life of the radioactive ^{140}La nuclei.

(3) The value of the ^{140}La half-life presented in this paper is

$$T_{1/2} = 1.6808(18) \text{ days}$$

$$\text{or } \lambda = 0.47749(20) \times 10^{-5} \text{ s}^{-1}.$$

This value is in good agreement with earlier measurements as given in Table 1. The measurement with semiconductor detectors in a wide range of counting rates and large dead times yields correct results when proper consideration of all necessary corrections is taken care off.

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