REFERENCE SOURCES FOR THE CALIBRATION OF THE AUTOCORRELATION SINGLE-CRYSTAL SCINTILLATION TIME SPECTROMETER

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A set of nine radioactive sources were measured for calibration of the autocorrelation single-crystal scintillation time spectrometer (ASCSTS) by using the delayed coincidences method. The measurements were performed in a wide time range, from 4 ns to several hundreds µs, and for energies starting from ~ 5 keV. The focus of the present paper was to measure the low energy threshold of the nuclear radiation detection, to determine the measurable time range with ASCSTS and to check the method sensitivity.

Key words: Radioactive sources, autocorrelation single-crystal scintillation time spectrometer (ASCSTS).

1. INTRODUCTION

Nano- and microsecond short-lived nuclear states arising from the radioactive decay are identified in most cases with the delayed coincidence method, usually with two detectors recording excitation and de-excitation of an isomeric state. The number of true coincidences is \( N_{\text{true}} \sim \omega_1 \omega_2 \), where \( \omega_1 \) and \( \omega_2 \) are the solid angles corresponding to the angles at which detectors are placed; therefore, to provide the maximum true coincidence recording efficiency, the solid angle for both detectors should be \( \omega = 4\pi \). The simplest way to meet this requirement is to use a single-crystal coincidence spectrometer, with the radioactive source implanted or placed in the sensitive volume of a scintillator. The single-crystal scintillation time spectrometer method, the experimental arrangement and electronics have been described in detail earlier [1–3] and will only be discussed briefly in the present paper.

The autocorrelation single-crystal scintillation time spectrometry (ASCSTS) method is a version of the delayed coincidence method. The method is characterized by a high efficiency of recording coincidences resulting from the use of only one
detector that records the radiation emitted from the nucleus in a geometry close to 4π. This method allows the measurements of the half-lives for excited and ground states, in the range of 4 ns to several hundreds µs. Since plastic scintillators with short decay times provides almost 100% detection efficiency of the charged particles, the low-energy limit for detecting radiation is defined only by the electronic noise of the photomultiplier. The three-dimensional E-T-E2 mode allows energy spectra corresponding to transitions exciting and de-exciting the isomeric states to be recorded in addition to the time spectrum. This demonstrates that the autocorrelation single-crystal time spectrometer can be effectively used to search for nuclear isomeric states.

2. CHARACTERISTICS OF THE STANDARD SOURCES

Since the ASCSTS technique involves analysis of energy and time correlations of nuclear radiation, it is necessary to adjust electronics in a proper way in order to achieve the optimum measurement conditions: the lowest energy threshold for the energy of the nuclear radiation that can be recorded by a detector, and a measurement of the minimal and maximal lifetimes accessible with this method. To achieve these conditions, the level of photomultiplier noise, the scintillator decay time, the pulse duration from the photomultiplier, type of the plastic scintillator and the discriminator dead time should be considered. In the present measurements, ASCSTS was based on a XP2020 photomultiplier type, a NE104 plastic scintillator type with decay time of τ = 1.9 ns and a discriminator ORTEC CFD-584. A set of standard sources that allows the recording of delayed coincidences to determine time and energy range was employed in the present work. Table 1 shows the data characterizing the standard sources. The set consists of 57Co, 73As, 90Sr, 153Gd, 169Yb, 225Ac, 226Ra, 232Th and 241Am, including also the products of their decay.

Table 1

<table>
<thead>
<tr>
<th>Source</th>
<th>T½ source</th>
<th>Measured nucleus</th>
<th>Level energy (keV)</th>
<th>T½ of level (ns)</th>
<th>E1 (keV)</th>
<th>E2 (keV)</th>
<th>Cascade (% per decay)</th>
</tr>
</thead>
<tbody>
<tr>
<td>57Co</td>
<td>271.8 d</td>
<td>57Fe</td>
<td>14.4</td>
<td>98.1(3)</td>
<td>Eγ = 122.1</td>
<td>Eγ = 14.4</td>
<td>85.6</td>
</tr>
<tr>
<td>73As</td>
<td>80.3 d</td>
<td>73Ge</td>
<td>13.2</td>
<td>2940(30)</td>
<td>Eγ(53.5) = 42.4</td>
<td>Eγ(13.2) = 11.8</td>
<td>100</td>
</tr>
<tr>
<td>90Sr</td>
<td>2.89 y</td>
<td>90Zr</td>
<td>1760.7</td>
<td>61.3(25)</td>
<td>Eβ = 2280</td>
<td>Eγ = 1758.2</td>
<td>0.0115</td>
</tr>
<tr>
<td>153Gd</td>
<td>241.6 d</td>
<td>153Gd</td>
<td>103.2</td>
<td>3.90(3)</td>
<td>Eγ(69.7) = 21.2</td>
<td>Eγ(103.2) = 55.3</td>
<td>15</td>
</tr>
<tr>
<td>169Yb</td>
<td>32.0 d</td>
<td>169Tm</td>
<td>316.2</td>
<td>66(7)</td>
<td>Eγ(63.1) = 53.0</td>
<td>Eγ(198) = 138.6</td>
<td>96</td>
</tr>
<tr>
<td>225Ac</td>
<td>10.0 d</td>
<td>213Po</td>
<td>0</td>
<td>3700(30)</td>
<td>Eα = 1422</td>
<td>Eγ = 8376</td>
<td>98</td>
</tr>
<tr>
<td>226Ra</td>
<td>1600 y</td>
<td>212Po</td>
<td>0</td>
<td>143.5(33) µs</td>
<td>Eγ = 3270</td>
<td>Eγ = 7833</td>
<td>100</td>
</tr>
<tr>
<td>232Th</td>
<td>1.4x1010 y</td>
<td>222Po</td>
<td>0</td>
<td>298(3)</td>
<td>Eγ = 2270</td>
<td>Eγ = 8784</td>
<td>64</td>
</tr>
<tr>
<td>241Am</td>
<td>443 y</td>
<td>237Np</td>
<td>59.5</td>
<td>68.3(2)</td>
<td>Eα = 5389; 5544</td>
<td>Eα = 59.5 = 37.4</td>
<td>~ 100</td>
</tr>
</tbody>
</table>
where $E_1$ and $E_2$ are the energy values for the radiation populating (in brackets) and depopulating the isomeric state, respectively; $E_K$, $E_L$, and $E_\beta$ characterize the energy of conversion electrons and the energy limit for $\beta$ particles; $E_\alpha$ specifies the energy range of the recorded $\alpha$ particles.

In most cases the radioactive sources were prepared in the form of a “sandwich”: the radioactive liquid was dropped on the plastic scintillator and dried. On the scintillator surface a thin layer of dichloroethane was deposited and then covered with another plastic scintillator similar in shapes and dimensions with the original one. Produced on styrene base, the dichloroethane dissolved the surface layers of the scintillator, thus providing the $4\pi$-geometry necessary for recording the nuclear radiation.

3. DETERMINATION OF THE LOW ENERGY THRESHOLD FOR THE NUCLEAR RADIATION DETECTION

The energy calibration of a scintillation detector with plastic scintillator that records charged particles and gamma-rays presents some difficulties due to the poor energy resolution. To solve this problem we proceed as described below: we recorded the proper time distribution corresponding to the lifetime of the isomeric state and then increased the discrimination level up to a value for which the acquisition of the exponential distribution stops. This level corresponds to the minimum energy of the electromagnetic transition from the isomeric cascade than can be recorded.

$^{241}\text{Am}$

The 59.9 keV isomeric state with a lifetime $T_{1/2}=67(2)\text{ ns}$, fed $\sim 100\%$ by $\alpha$-decay of $^{241}\text{Am}$ to $^{237}\text{Np}$ is a very favorable case for a quick calibration of the scintillation spectrometer with a plastic scintillator. The energy of the conversion electrons from the 59.5 keV transition which does not exceed the energy $E_L=37.4\text{ keV}$ allows the extrapolation of the energy calibration in the low energy range. Fig. 1 shows the decay curve for the 59.5 keV state in $^{237}\text{Np}$ measured with this source and the measured value for this transition is $T_{1/2} = 68.3(2)\text{ ns}$. Fig. 2 shows the energy spectrum of $^{241}\text{Am}$ decay, and also the spectra of radiation populating and depopulating the isomeric state in $^{237}\text{Np}$.

$^{73}\text{As}$

The use of $^{73}\text{As}$ isotope was necessary to determine the low-energy threshold for detecting the nuclear radiation. Fig. 3 displays the result of the half-life measurement of the isomeric state 13.2 keV from $^{73}\text{Ge}$ ($T_{1/2} = 2.94(3)\text{ \mu s}$). Fig. 4 shows the energy spectra of radiations emitted in the $^{73}\text{As} \rightarrow ^{73}\text{Ge}$ decay, obtained with the ASCSTS in the present experiment.
Fig. 1. Half-life of the 59.5 keV state in $^{237}$Np.

Fig. 2. (1) The radiation spectrum for the $^{241}$Am decay; (2) Radiation spectrum populating the isomeric states in $^{237}$Np; (3) Radiation spectrum depopulating the isomeric states in $^{237}$Np.
Fig. 3. Half life measurement in $^{73}$Ge.

Fig. 4. (1) The radiation spectrum for the $^{73}$As decay; (2) Radiation spectrum populating the isomeric states in $^{73}$As; (3) Radiation spectrum depopulating the isomeric states in $^{73}$As.
Another candidate for determining the low energy threshold for detection of the gamma-rays is $^{57}$Co. The measurements of the half-life of the 14.4 keV in $^{57}$Fe were performed in a geometry closed to $2\pi$ with a plastic scintillator of $25 \times 25$ mm. The radioactive source was obtained by irradiating a natural iron foil (4 µm thickness) with a proton beam at the cyclotron accelerator of NIPNE-HH, Bucharest. The radioactive source was measured in two ways: first, the source was placed at 1 mm from the surface of the plastic scintillator (curve 1 in Fig. 5) and, second, a polyethylene filter was placed between the source and plastic detector (curve 2 in Fig. 5).

![Graph](attachment:image.png)

Fig. 5. Half life measurement of the 14.4 keV in $^{57}$Fe. (1) Measurement performed without the filter; (2) Measurement done with the plastic filter. (3) $^{137}$Cs – reference source (random coincidences).

The conversion electrons from the 14.4 keV energy transition were completely absorbed by the thick polyethylene foil (1 mm thick) filter [4]. Although a strong absorption of $\gamma$-rays having 14.4 keV energy takes place (the absorption coefficient
equal to three), both curves have the same shape with a lower counting rate in the second case. The existence of the exponential distribution due to the lifetime of the 14.4 keV isomeric state from $^{57}\text{Fe}$ shows that if in the first case the recording of the isomeric cascade takes place by recording conversion electrons, gamma-rays with an energy of 14.4 keV, X-ray and Auger electrons, in the second case the exponential distribution is only due to gamma-rays with an energy of 14.4 keV.

### 4. DETERMINATION OF THE TIME RANGE WITH ASCSTS

The measurements of the lifetimes of the isomeric nuclear states determined with ASCSTS ranges from a few ns to hundreds µs. The set of radioactive sources used covers almost entirely this time range. A special interest presents the determination of both the lower and upper limits of the lifetimes that are possible to study with ASCSTS.

#### $^{153}\text{Gd}$

The $^{153}\text{Gd}$ source was used to demonstrate the possibility of measuring nanosecond lifetime with the ASCSTS method. The $^{153}\text{Gd}$ was a product of the tantalum target splitting by fast protons of 660 MeV at the phasotron of LNP, JINR, Dubna. After the chemical separation from the group of rare earth elements, the Tb fraction was mass-separated in order to achieve an isobar chain with A = 153. After the mass-separation the source was embedded into aluminum foil and then placed into a plastic scintillator. Fig. 6 shows the results of measuring the half-life of the state with 103.18 keV (3.95(11) ns) from $^{153}\text{Eu}$ excited in the $^{152}\text{Gd}$ decay.

The dead time of the discriminator used in the measurements did not exceed 10 ns and the threshold set for recording radiation was lower than 30 keV. The lifetime measured for the state with energy of 103.18 keV from $^{153}\text{Eu}$ is the lower limit for time measurements reachable with this experimental setup (for the use of the fotomultiplier XP2020 – a pulse duration of 5 ns – coupled with the plastic scintillator NE104).

#### $^{226}\text{Ra}$

The reason to measure the radioactive source of $^{226}\text{Ra}$ was to show the upper limit of the time measurements reachable with ASCSTS. The half-life of the ground state of $^{214}\text{Po}$ was measured. The isotope $^{214}\text{Po}$ is a member of a long radioactive chain starting with $^{226}\text{Ra}$: $^{226}\text{Ra} \xrightarrow{\alpha} ^{222}\text{Rn} \xrightarrow{\alpha} ^{218}\text{Po} \xrightarrow{\alpha} ^{214}\text{Pb} \xrightarrow{\beta^-} ^{214}\text{Bi} \xrightarrow{\beta^-} ^{214}\text{Po} \xrightarrow{\alpha} ^{210}\text{Pb}$ and has a half-life $T_{1/2} = 157.6$ (33) µs. The result of the performed measurement is shown in Fig. 7.

In this way it was proved that the ASCSTS method (the particular setup – XP2020 photomultiplier and NE104 plastic scintillator) can be used for measuring a large range of life-times spanning more than four orders of magnitude, ranging
from \( \sim 4 \) ns to a few hundreds of \( \mu \text{s} \). The following radioactive sources \(^{169}\text{Yb} \), \(^{225}\text{Ac} \) and \(^{232}\text{Th} \) were measured for a complete study along the entire time range achievable with ASCSTS and to demonstrate the usefulness of the method for measuring lifetimes in nuclei belonging to long radioactive chains.

Fig. 6. Half life of the 103.18 keV state in \(^{153}\text{Eu} \).

\[
T_{1/2} = 3.95(11) \text{ ns}
\]

Fig. 7. Half life of the \(^{214}\text{Po} \) measured in \( \alpha \)-decay to \(^{210}\text{Pb} \).
The results of the half-life measurements of the 316.15 keV excited state in $^{169}$Tm, ($T_{1/2} = 661(7)\text{ ns}$) are shown in Fig. 8. Low energy radiation was recorded with a plastic scintillator (25x25 mm) in $2\pi$-geometry applying an energy threshold $E \geq 30\text{ keV}$. The $^{169}$Yb source was prepared in the same way as the $^{153}$Gd source.

$^{169}$Yb

The half-lives of the ground states of the 213Po and 212Po were measured using a source of a complex isotopic composition. 213Po and 212Po are two members in the radioactive chain beginning with $^{225}$Ac and $^{232}$Th respectively.

In the first case, the source was extracted from the $^{225}$Ac source by the ion-exchange method and dried on a plastic scintillator. The observed time distribution with $T_{1/2} = 3.65\mu$s was assigned to the 213Po ground state decay [5].

To determine the ground state half-life of 212Po the source was prepared as follows: an aqueous solution of thorium nitrate was deposited onto the surface of a plastic scintillator (6 mm thick with a 10 mm diameter) and dried. The scintillator surface was covered with styrene and bond together to another scintillator of the same type and shape to ensure the $4\pi$ geometry. Around 10 mg of thorium were applied [6]. Fig. 8 displays the time spectra characterizing both the 213Po and 212Po half-lives.
5. SENSITIVITY OF THE METHOD

Besides the key features of the method, a low threshold for radiation energy and a long time range for life-time measurements, another important issue is to demonstrate that the ASCSTS method has a good sensitivity for recording the delayed coincidences. The method sensitivity depends on several factors: the efficiency of the spectrometer to record the radiation, the detection solid angle and the measurement time interval.

Using 4π geometry gives the ability to study the low intensity isomeric cascades. The recording efficiency of the delayed coincidences can be evaluated in two ways: by using the experimental exponential time distribution or by evaluating the intensity of the delayed gamma-rays with an additional HPGe detector along with the ASCSTS spectrometer. To prove the method sensitivity (for this particular setup) two measurements were performed for $^{90}\text{Sr}$ and $^{229}\text{Th}$.

$^{90}\text{Sr}$

The first measurement, shown in Fig. 9, verifies the possibility to measure the life-time of an isomeric cascade which is de-excited through a very weak transition. The 1760.7 keV isomeric state ($T_{1/2} = 61.3$ ns) is populated in beta decay of $^{90}\text{Y}$ (coming from $\beta$-decay of $^{90}\text{Sr}$) and is de-excited through an E0 transition with the energy of 1760.7 keV and an intensity of 0.0115% to the ground state of $^{90}\text{Zr}$.

![Fig. 9. Time spectra for the decay of $^{90}\text{Sr}$. The level of random coincidences is shown by the decay of $^{60}\text{Co}$.](image)

$^{229}\text{Th}$

The second measurement shows the intensity of the delayed gamma rays emitted in beta decay of $^{213}\text{Bi}$ which is a member of the long radioactive chain starting with $^{229}\text{Th}$. We used a semiconductor HPGe detector (without any shielding) with 10% efficiency. The experiment with HPGe detector and a $^{229}\text{Th}$
source was 22 hours long. The ASCSTS counting rate was ~1300 counts/s and that of the HPGe detector (source + background) was ~170 counts/s (80 counts/s + 90 counts/s). The coincidence spectrum features distinct γ-lines coming from the $^{213}\text{Bi} - ^{213}\text{Po}$ decay [5] (Fig.10). These lines are distinct against a very complex γ-spectrum from the decay of 12 nuclei ($^{229}\text{Th}$ chain) and from the background radiation with the admixtures from the radioactive $^{232}\text{Th}$ chain nuclei and $^{226}\text{Ra}$, $^{137}\text{Cs}$, $^{60}\text{Co}$ and $^{40}\text{K}$ nuclei. Table 2 presents experimental intensities belonging to the β-decay of $^{213}\text{Bi}$ in accordance with well-known data [6].

![Gamma spectrum for the coincidences of the $^{213}\text{Bi}$ decay.](image)

### Table 2

<table>
<thead>
<tr>
<th>$E_\gamma$ (keV)</th>
<th>$I_\gamma$ (%) [7]</th>
<th>$I_\gamma$ (%) [5]</th>
<th>$S(\Delta S)$</th>
<th>$S/\varepsilon \times 10^3$</th>
<th>$I_\gamma$ (%) (present experiment)</th>
</tr>
</thead>
<tbody>
<tr>
<td>292.76</td>
<td>0.42(2)</td>
<td>0.39(2)</td>
<td>655(64)</td>
<td>81(8)</td>
<td>0.31(4)</td>
</tr>
<tr>
<td>440.43</td>
<td>26.1(3)</td>
<td>24.6(15)</td>
<td>35070(195)</td>
<td>6810(38)</td>
<td>26.1(2)</td>
</tr>
<tr>
<td>659.77</td>
<td>0.036(2)</td>
<td>0.44(3)</td>
<td>30(12)</td>
<td>9(3)</td>
<td>0.035(11)</td>
</tr>
<tr>
<td>710.81</td>
<td>0.010(1)</td>
<td>0.0119(10)</td>
<td>12(7)</td>
<td>4(2)</td>
<td>0.015(8)</td>
</tr>
<tr>
<td>807.38</td>
<td>0.241(15)</td>
<td>0.238(18)</td>
<td>185(16)</td>
<td>70(6)</td>
<td>0.27(2)</td>
</tr>
<tr>
<td>867.98</td>
<td>0.011(1)</td>
<td>0.0123(11)</td>
<td>15(8)</td>
<td>6(4)</td>
<td>0.023(13)</td>
</tr>
<tr>
<td>1003.55</td>
<td>0.050(5)</td>
<td>0.053(3)</td>
<td>23(6)</td>
<td>11(3)</td>
<td>0.04(1)</td>
</tr>
<tr>
<td>1110.02</td>
<td>0.259(16)</td>
<td>0.251(17)</td>
<td>115(11)</td>
<td>59(6)</td>
<td>0.23(2)</td>
</tr>
<tr>
<td>1119.29</td>
<td>0.050(3)</td>
<td>0.051(3)</td>
<td>20(5)</td>
<td>11(3)</td>
<td>0.04(1)</td>
</tr>
</tbody>
</table>

“S” is the number of counts and $\varepsilon$ is the recording efficiency coefficient of the HPGe detector.
6. CONCLUSIONS

Thus the set of radioactive sources with known transition energies and half-lives of nuclear isomeric states made possible the time and energy calibration of the spectrometer. The present paper proves that by employing the ASCSTS method an energy calibration can be performed down to 5 keV. Also, the time calibration can be done in the interval ranging from 4 ns up to at least 200 µs.

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