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# A spectrometer for lifetime determination by $\beta-\gamma-\gamma$ delayed coincidence technique at KUR-ISOL

Y. Kojima [a,*,1], H. Hayashi [b,2], M. Shibata [b], S. Endo [a], K. Shizuma [a], A. Taniguchi [c]

[a] Graduate School of Engineering, Hiroshima University, Higashi-Hiroshima 739-8527, Japan
[b] Radioisotope Research Center, Nagoya University, Nagoya 464-8602, Japan
[c] Research Reactor Institute, Kyoto University, Kumatori 590-0494, Japan

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ABSTRACT

A new spectrometer for measuring nuclear level lifetimes has been installed at the on-line isotope separator of the Kyoto University Reactor. The spectrometer consists of a $LaBr_3$(Ce), Ge and a thin plastic scintillation detector, and the lifetimes are determined using a $\beta-\gamma-\gamma$ delayed coincidence technique. In this study, the $LaBr_3$ detector was used to obtain time spectra, whereas the Ge detector was used to select a desired $\gamma$ branch. The energy dependence of the time resolutions was measured down to a photon energy of 100 keV. The lifetimes measured for the excited levels in $^{93}$Sr and $^{148}$Ce agree well with their evaluated values. The lifetime of 8.5(5) ns was obtained for the first time for the 98.2 keV level in $^{148}$Pr.



## 1. Introduction

Extensive spectroscopic studies have been conducted on the fission products of $^{235}$U. Among these products, nuclei in the rare earth region having a mass number of around 150 still attract continuing attention because they provide an opportunity to study the transition from spherical nuclei to the quadrupole–octupole deformed nuclei. For example, Hayashi et al. recently reported that the experimental two-neutron separation energies for $^{147\text{-}149}$La deviate from their theoretically evaluated values [1,2]. This systematic deviation strongly suggests that a change in the nuclear structure which is not taken into account in the theoretical mass evaluation occurs. To investigate the nuclear structure of $^{147\text{-}149}$La and their neighboring nuclei, we are now planning to perform lifetime measurements at the on-line isotope separator installed at the Kyoto University Reactor (KUR-ISOL) [3]. In these experiments, a spectrometer consisting of a thin plastic scintillation detector and a recently developed $LaBr_3$(Ce) scintillator [4–6] will be used to determine the lifetimes by a $\beta-\gamma$ delayed coincidence method. As listed in Table 1, $LaBr_3$, among all the scintillation materials, shows the best energy resolution and a moderate detection efficiency for $\gamma$-rays. Table 1 also shows that the $LaBr_3$ scintillator has a good decay time of 16 ns, whereas those of most of the inorganic scintillators are a few hundred nanoseconds or longer, except for $BaF_2$. A short decay time reduces variance of emission timing signals of scintillation photons. Thus, the $LaBr_3$ scintillator has a good time resolution and is used for measuring the lifetimes [8–12].

In this work, we study the properties of our spectrometer installed at KUR-ISOL as well as its stability during long-term measurements. Next, we apply this spectrometer to measure lifetimes of excited levels of mass-separated $^{93}$Sr and $^{148}$Ce nuclides. By this experiment, we demonstrate the feasibility of lifetime determination, in the range of nanoseconds, by means of a slope method. Finally, the lifetime of the 98.2 keV level in $^{148}$Pr was measured, for which no experimental data is yet available.

## 2. Details of the spectrometer

### 2.1. $LaBr_3$ detector

The $LaBr_3$ detector used in this study is a commercially available detector (Canberra, model LABR). The crystal, which is 1.5-in. in diameter and 1.5-in. in thickness, was coupled with the Hamamatsu 14-pin photomultiplier tube (PMT) R6231. The $LaBr_3$ scintillator enclosed by a 0.7-mm-thick Teflon reflector was

* Corresponding author. Tel.: +81 52 789 2570; fax: +81 52 789 2567.
E-mail address: kojima.yasuaki@f.mbox.nagoya-u.ac.jp (Y. Kojima).
[1] Present address: Radioisotope Research Center, Nagoya University, Nagoya 464-8602, Japan.
[2] Present address: Institute of Health Biosciences, The University of Tokushima Graduate School, Kuramoto, Tokushima 770-8509, Japan.

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**Table 1**
Properties of scintillation materials [4]. Detection efficiencies relative to NaI are calculated for a 1.5 in. × 1.5 in. crystal using the Geant4 computer code [7].

| Scintillation material | Energy resolution for 662 keV γ-ray (typical) (%) | Decay time (ns) | Detection efficiency for 1 MeV γ-ray |
|---|---|---|---|
| $LaBr_3$ | < 3 | 16 | 1.5 |
| $BaF_2$ | 12 | 0.6 | 1.6 |
| BGO | 12 | 300 | 4.0 |
| NaI | 7 | 250 | 1 |

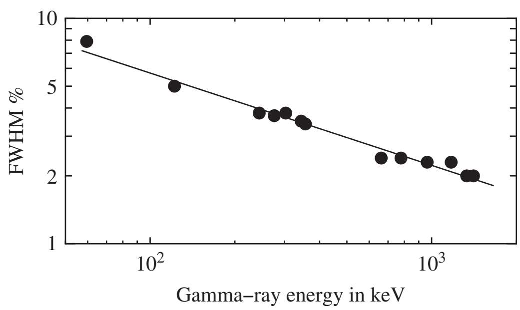


**Fig. 1.** Energy resolution (FWHM) of the $LaBr_3$ detector observed at an applied voltage of 800 V.

placed in a 0.5-mm-thick aluminum housing. The R6231 tube is optimized for measuring the pulse height and not for measuring time. Thus, its timing property would not be as good as that of a faster PMT. However, the R6231 PMT is expected to have a good energy resolution, and is therefore suitable for application to experiments using the mass separator, because fission products usually show complex γ spectra. The output signal from the anode was directly connected to a timing module described later. The energy spectra were obtained from the dynode signal, through a charge sensitive preamplifier and a main amplifier. The operating bias voltage specified by the manufacturer was 600–800 V. Hence, before the timing measurements, the energy resolutions and pulse heights were examined in this voltage range using standard radioactive sources such as $^{241}$Am, $^{133}$Ba, $^{152}$Eu, $^{137}$Cs, and $^{60}$Co. A shaping time of 2 μs of the main amplifier was used for this measurement.

Fig. 1 shows the energy resolution of the $LaBr_3$ detector obtained at an applied voltage of 800 V. An energy resolution (FWHM) of 2.4% was obtained for a 662 keV γ-ray. Energy resolution was also measured at bias voltages of 600 and 700 V. No difference was observed in the shapes of the γ-ray peaks, as expected. However, amplitudes of signals from the PMT varied significantly in this voltage range, that is, the signal height increased as the applied voltage increased, as expected. When we used electronic devices having an input impedance of 50 Ω, the anode signals obtained at the 600 V bias voltage were less than 20 mV for X-rays ( $\leq$ 100 keV). These small anode signals were below the minimum discrimination level of the timing modules. Therefore, the bias voltage of 800 V was applied for subsequent experiments.

The background spectrum was also measured in a 5-cm-thick lead shielding surrounding the $LaBr_3$ detector. $LaBr_3$ contains an abundance of the naturally occurring radioisotopes $^{138}$La, $^{227}$Ac and its daughter nuclides, so that a significant self-background was observed. The background rate of this crystal was 62 cps in the 35–3000 keV region and 37 cps in the 35–1500 keV region. The Ba K,L,M, ... X rays sequentially emitted after the β decay of $^{138}$La were summed in the crystal, so that the peak was observed at 37 keV, which corresponds to the K binding energy of Ba atom. The counting rate of the Ba X-ray sum-peak was 18 cps.

*2.2. Timing properties of the spectrometer*

Our system for measuring the lifetimes is based on the β−γ delayed coincidence technique. The $LaBr_3$ detector described earlier is used for γ-ray measurements. Beta-rays are detected using a thin plastic scintillation detector. A 1-mm-thick plastic scintillator (pilot-U, 35 × 35 mm) was coupled to a fast PMT assembly (Hamamatsu H2431-51), and it was light-shielded by a 41-μm-thick reflection film. The detection efficiency was calculated using the Geant4 computer code [7]. The efficiency obtained at a source-to-detector distance of 25 mm was approximately 0.5% for the β−γ coincidence measurements, assuming that the β transitions have a maximum energy of 2 MeV and the γ-rays have an energy of 100 keV.

The electronic unit used in this experiment consisted of standard nuclear instrumentation modules (NIM): two constant-fraction discriminators (CFDs, Canberra 2126) and a time-to-amplitude converter (TAC, Ortec 567). A 2-m-long delay cable is used to set the constant-fraction timing for the $LaBr_3$ detector, and a 0.25-m-long cable is used for the plastic scintillator. The lengths correspond to approximately 80% of the fall time of the anode signals from each detector. A β signal started the functioning of the TAC unit. The energy and TAC signals are recorded in list mode. The measuring system was calibrated using the Ortec 462 time calibrator. From the calibration measurements, a time calibration curve which shows a relation between a channel number and a time was obtained. This curve showed a good linearity, and the deviation of each data point from the calibration line was less than 20 ps.

Prompt time spectra were measured using standard radioactive sources, $^{60}$Co and $^{134}$Cs, to evaluate the timing properties of this system. The $^{60}$Co decay ($Q_\beta = 2824$ keV) is characterized by its simple decay scheme [13]. It decays to a level of 2506 keV in $^{60}$Ni (lifetime $\tau$ of 0.43 ps), and 1173 and 1333 keV γ-rays are emitted via the 1333 keV level ($\tau = 1.0$ ps). The $^{134}$Cs nuclide β-decays to $^{134}$Ba ($Q_\beta = 2059$ keV) and emits γ-rays [13]. The energies of the intense γ-rays are 563, 569, 605, 796, and 802 keV. It should be noted that the 1643 keV level in $^{134}$Ba has a lifetime of 112 ps. However, the lifetime is shorter than the expected time resolution of the $LaBr_3$ detector (a few hundred picoseconds). In addition, the β feeding intensity to this level is less than 2.5%. Therefore, the effect of the 112 ps lifetime on the measurement of the prompt time peak is small and hence can be neglected.

To estimate the timing properties in the energy region lower than 100 keV, a method using an X-ray converter was also applied [14]. In this method, an appropriate target such as tungsten is placed near a radioactive source. The γ-rays from the source excite the target atoms, and the X-rays are emitted from the target after a short time (a time span range of nanoseconds). In order to obtain time spectra down to a photon energy of 44 keV, we used the following materials as converters: Tb ($Tb_4O_7$, powder, 5.1 g), Yb ($Yb_2O_3$, powder, 5.5 g), W and Pb (0.5-mm-thick metallic sheet, 20 × 20 mm).

The prompt time peaks were also measured using short-lived $^{93}$Rb and $^{148}$Ce to obtain the timing properties in the energy region of 100–500 keV. The $^{93}$Rb and $^{148}$Ce nuclides were obtained as mass-separated beams from the fission products of $^{235}$U using the on-line isotope separator KUR-ISOL [3]. In this experiment, γ-rays were also measured using a Ge detector to select the desired γ branch. Further details of the source preparation of $^{93}$Rb and $^{148}$Ce are described in Section 3.1.

The insets of Fig. 2 show the prompt time spectra obtained after careful adjustment of the time walks of the CFDs. The prompt time spectra were obtained by off-line sorting. In this sorting, gates were set on the full-energy γ peaks detected with the $LaBr_3$ detector. Here, a γ-ray peak observed around 99 keV using the $LaBr_3$ detector was a triplet: 98.0, 98.2 and 99.0 keV