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The Generalized Centroid Difference method for lifetime measurements via γ-γ coincidences using large fast-timing arrays

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Abstract. A novel method for direct electronic "fast-timing" lifetime measurements of nuclear excited states via γ-γ coincidences using an array equipped with N very fast high-resolution LaBr$_3$(Ce) scintillator detectors is presented. The generalized centroid difference method provides two independent "start" and "stop" time spectra obtained without any correction by a superposition of the $N(N-1)/2$ calibrated γ-γ time difference spectra of the N detector fast-timing system. The two fast-timing array time spectra correspond to a forward and reverse gating of a specific γ-γ cascade and the centroid difference as the time shift between the centroids of the two time spectra provides a picosecond-sensitive mirror-symmetric observable of the set-up. The energy-dependent mean prompt response difference between the start and stop events is calibrated and used as a single correction for lifetime determination. These combined fast-timing array mean γ-γ zero-time responses can be determined for 40 keV $< E_\gamma <$ 1.4 MeV with a precision better than 10 ps using a ^{152}Eu γ-ray source. The new method is described with examples of (n,γ) and (n,f,γ) experiments performed at the intense cold-neutron beam facility PF1B of the Institut Laue-Langevin in Grenoble, France, using 16 LaBr$_3$(Ce) detectors within the EXILL&FATIMA campaign in 2013. The results are discussed with respect to possible systematic errors induced by background contributions.

1 Introduction

As an essential nuclear observable, the lifetimes of excited states are needed to determine the reduced transition probabilities which are used to test the model dependent intrinsic structure of the nuclear excited states. The electronic fast-timing technique in combination with very fast scintillator detectors is picosecond sensitive [1–3] and therefore is capable of overlapping with complementary techniques such as the Recoil Distance Method [4] and Coulomb excitation [5]. For the picosecond regime, the fast-timing technique is based on the determination of centroids of time distributions (first moment of a time spectrum [6]) generated as time difference spectra of consecutive γ-γ transitions measured using two start and stop γ-ray detectors. Assuming no background contribution, the experimentally obtained "delayed" time distribution $D(t)$ is a convolution of the Prompt Response Function (PRF) of the setup $P(t)$

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with an exponential decay as:

$$D(t) = n\lambda \int_{-\infty}^{t} P(t' - t_0)e^{-\lambda(t - t')}dt' \quad \text{with} \quad \lambda = 1/\tau, \tag{1}$$

where n is the total number of detected γ-γ events in the time difference spectrum, λ the transition (decay) probability and τ the mean lifetime of the nuclear excited state interconnected by the γ-γ cascade and t_0 is the position (centroid) of the PRF $P(t)$. The experimental PRF provides important information on timing characteristics and is obtained for lifetimes which are smaller than 1 ps (systematic errors are expected to be larger). For lifetimes which are larger than the FWHM of the PRF, the mean lifetime is obtained directly using the slope method [1]. Lifetimes which are smaller than the FWHM of the PRF can be determined using the centroid-shift method [6]. For the case of a Gaussian PRF with standard deviation $\sigma \cong \text{FWHM}/2.355$, the pure statistical time resolving power δt of a two detector timing system is given by:

$$\delta t = \frac{\sigma}{\sqrt{n}}. \tag{2}$$

The centroid or center of gravity C^D is the first moment of the statistical time distribution:

$$C^D = < t > = \frac{\int_{-\infty}^{\infty} tD(t)dt}{\int_{-\infty}^{\infty} D(t)dt}, \quad \delta C^D = \delta t = \sqrt{< t^2 > - < t >^2}, \tag{3}$$

where $D(t)$ is given by Eq. (1). According to the centroid-shift method, the centroid of a delayed time spectrum is displaced by the mean lifetime from the centroid of its convoluted PRF:

$$\tau = C^D_{\text{stop}} - C^P_{\text{stop}} \tag{4}$$

or if the functions of the two detectors are interchanged to obtain the "anti-delayed time spectrum" [1],

$$\tau = C^P_{\text{start}} - C^D_{\text{start}}, \tag{5}$$

where C^P is the "prompt centroid" of the PRF. The subscript "start" ("stop") indicates that the decay transition with its lifetime information provided the start (stop) timing signal of the two detector timing system.

The centroid-shift method is in principle very simple since the mean lifetime is directly obtained from the centroids of the delayed and the prompt time spectrum. In addition, the centroid-shift method is independent of the shape of the PRF, as demonstrated experimentally in Refs. [7, 8]. The major problem in γ-γ fast-timing experiments is that the prompt centroid is dependent on the time response, i.e. the physical zero-time versus energy relation $t_0(E_\gamma)$ (the time-walk characteristics) of both detectors of the γ-γ fast-timing setup, thus $C^P = C^P(E_{\text{start}}, E_{\text{stop}}) = t_0(E_{\text{start}}) + t_0(E_{\text{stop}})$. The calibration of the detector time response is possible, however, the analysis of an N detector timing system becomes complex for $N \gg 2$ as the time response of each single detector has to be calibrated, whereby systematic errors can easily be introduced [9].

The aim of this work is to present the Generalized Centroid Difference (GCD) method, which was developed to provide high-accuracy centroid-shift measurements with an N γ-ray detector timing system by using a simple approach.

2 The Generalized Centroid Difference method

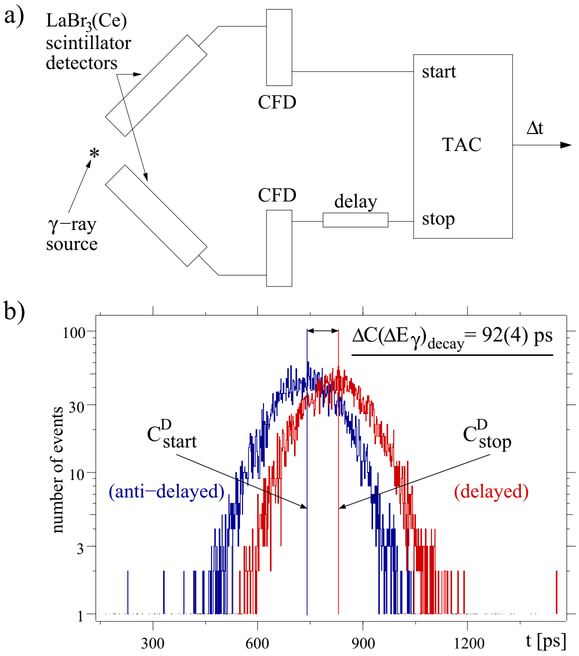

Figure 1. a: The standard electronic γ-γ fast-timing circuitry using Constant Fraction Discriminators (CFD) for picosecond sensitive time-difference measurements with a Time-to-Amplitude Converter (TAC). b: Two time spectra are obtained dependent on whether the decay transition provided a stop signal (the delayed time spectrum) or a start signal (anti-delayed).

We first consider the standard γ-γ fast-timing setup consisting of two (start and stop) detectors, as presented in Fig. 1a. As both the start and the stop detectors see the same γ-ray source, two time distributions are obtained in the off-line analysis by setting a gate (narrow energy window) on the full energy peak of the decay transition γ_{decay} of a nuclear excited state interconnected by a specific γ_{feeder}-γ_{decay} cascade once by using the start detector and once by using the stop detector, while the feeding γ ray is detected by the other detector. Assuming no background contributions and according to Eqs. (4) and (5), the time difference between the centroids of the two time spectra presented in Fig. 1b corresponds to:

$$\begin{aligned} \Delta C(\Delta E_\gamma)_{\text{decay}} &= C^D(\Delta E_\gamma)_{\text{stop}} - C^D(\Delta E_\gamma)_{\text{start}} \\ &= \text{PRD}(\Delta E_\gamma)_{\text{decay}} + 2\tau, \end{aligned} \tag{6}$$

where $\Delta E_\gamma = E_{\text{feeder}} - E_{\text{decay}}$ is the energy difference of the two γ-rays of the cascade and $\text{PRD}(\Delta E_\gamma)_{\text{decay}} = C^P(\Delta E_\gamma)_{\text{stop}} - C^P(\Delta E_\gamma)_{\text{start}}$ is the Prompt Response Difference which describes the linearly combined γ-γ zero-time response of the two detector timing system. With respect to a start-stop inversion, the centroid difference is mirror symmetric. For the prompt case, this is equivalent to a hypothetical feeder-decay inversion of the cascade [3], thus:

$$\Delta C(\Delta E_\gamma)_{\text{decay}} = -\Delta C(-\Delta E_\gamma)_{\text{feeder}}, \tag{7}$$

$$\text{PRD}(\Delta E_\gamma)_{\text{decay}} = -\text{PRD}(-\Delta E_\gamma)_{\text{feeder}} \tag{8}$$

and accordingly:

$$\text{PRD}(\Delta E_\gamma = 0) = 0 \text{ and } |\Delta C(\Delta E_\gamma = 0)| = 2\tau. \tag{9}$$

Eqs. (6) to (9) represent the Mirror Symmetric Centroid Difference (MSCD) method for a two detector timing system. Due to linear combination of start and stop events, the MSCD method reduces possible systematic errors and cancels the typical systematic error by long term shifts due to drifts in the electronics that can be induced using centroid-shift measurements where the reversed gating is not used. The PRD mirror symmetry, Eq. (8), provides additional data points for a precise calibration of the PRD curve, PRD(E_γ), and makes the determination of the PRD for any energy combination ΔE_γ possible using [3, 8]:

$$\text{PRD}(E_{\text{feeder}} - E_{\text{decay}}) = \text{PRD}(E_{\text{feeder}}) - \text{PRD}(E_{\text{decay}}). \tag{10}$$

The PRD for the energy combination of a γ-γ cascade is used as a single correction for the determination of the lifetime according to Eq. (6) and therefore provides the sole uncertainty of the MSCD method, assuming no background contributions to the time spectra.

For the case of a fast-timing array with N almost equal fast-timing detectors and suitable electronics set-up (e.g. the one shown in Ref. [9]), the centroid differences ΔC_{ij} of all unique detector-detector combinations ij with $j \neq i \in N$ could be measured to provide the mean value as:

$$\overline{\Delta C} = \frac{2\sum_{i=1}^{N-1}\sum_{j=i+1}^{N}\Delta C_{ij}}{N(N-1)} = \overline{\text{PRD}} + 2\tau. \tag{11}$$

Eq. (11) represents the Generalized Centroid Difference (GCD) method and denotes that the time difference between start events (decay transition detected by a start detector) and stop events are statistically distributed around $\overline{\Delta C}$ or $\overline{\text{PRD}}$ independent of the detector-detector combination. Experimentally, the mean centroid difference is equal to the time shift between the two fast-timing-array time spectra generated by a simple superposition of the $N(N-1)/2$ start (stop) time spectra "TAC$_{ij}$" [9] (projected time spectra of detector-detector combinations ij). This identical procedure called the GCD method provides a substantial simplification of fast-timing analyses on N detector timing systems as the tedious determination of N detector time responses is eliminated. The complexity of the data analysis is reduced to that of a two detector timing system. Analogous to the MSCD method, the mean PRD is used as the only correction which reduces a possible systematic error to a minimum by taking advantage of the mirror symmetric representation of the GCD method.

3 The GCD method for EXILL&FATIMA

The GCD method was developed especially for large fast-timing arrays such as the "FATIMA" γ-ray spectrometer made of $N = 16$ 5% Ce doped LaBr$_3$ detectors that was installed at the Institut Laue-Langevin (ILL) in Grenoble, France. This FATIMA spectrometer was combined with part of the EXILL (EXOGAM at ILL [10]) spectrometer consisting of 8 HPGe EXOGAM Clover detectors. Within the EXILL&FATIMA campaign, fast electronic timing measurements were performed in prompt γ-ray spectroscopy experiments using neutron-capture and neutron-induced fission reactions at the highly collimated high-flux cold-neutron beam facility PF1B [11, 12] of the ILL. Triple coincidences were used in which a Ge gate selects the cascade of γ rays to be measured by the LaBr$_3$ detectors. A detailed description of the EXILL&FATIMA set-up is given in Ref. [13]. In a preparatory work, the $N(N-1)/2 = 120$ LaBr$_3$-LaBr$_3$ coincidence spectra of the FATIMA set-up were investigated for artefacts such as inter-detector Compton scattering. Such cross-talk events with false timing information have been detected in 16 adjacent and 8 other LaBr$_3$-LaBr$_3$ detector combinations, those are excluded from the main data analysis [13].

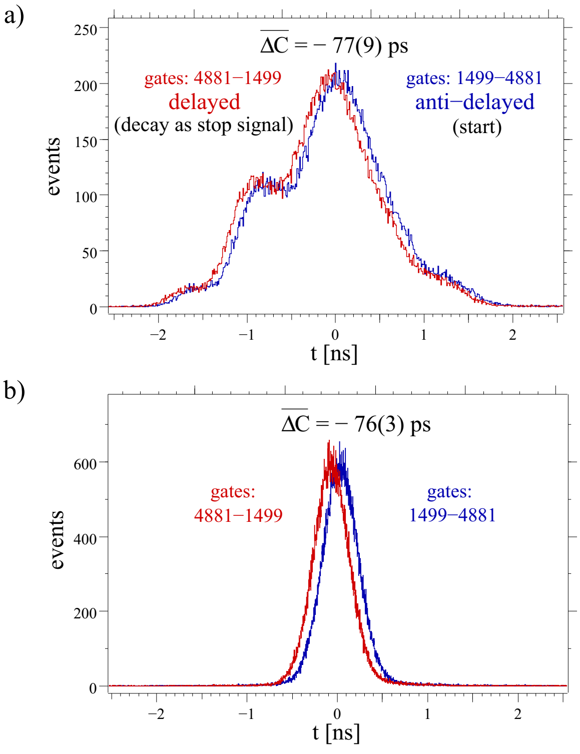

Figure 2. Principle of the GCD method. a: Superposition of 96 remaining time-difference spectra TAC$_{ij}$ of a prompt γ-γ cascade in ^{49}Ti with $\tau = 16(7)$ fs [14] as obtained from the 16 LaBr$_3$ fast-timing array of the EXILL&FATIMA spectrometer, by distinguishing between the stop events (decay transition detected as a stop signal) and the start events independent of the detector-detector combination ij. b: Result of the same data shown in Fig. 2a obtained after the alignment of the 96 TAC$_{ij}$ spectra using constant shift$_{ij}$ values (see the text for more detail) in order to improve the time resolution [FWHM=292(3) ps].

Fig. 2 illustrates the principle of the GCD method: The fast-timing data of the 96 unique detector-detector combinations (time spectra TAC_{ij}) are superimposed without any correction by distinguishing between the delayed (decay transition detected as stop signal) and the anti-delayed time spectrum of a γ-γ cascade to measure the centroid difference. In order to improve the precision of the measurement following Eq. (2), the time spectra TAC_{ij} are "aligned" using energy-independent constant shift_{ij} values, which means that both the start and stop events from the detector-detector combination ij are adjusted by the same shift_{ij} constant. As illustrated in Fig. 2b, this alignment does not introduce a systematic error and thus does not represent a correction, while it reduces the width of the distribution and thus the statistical error. These observations nicely confirm that the lifetime determination by means of centroid-shift measurements is independent of the shape of the PRF.

The major work of any fast-timing experiment is to calibrate the zero-time response of the set-up. Fig. 3 shows the (mean) PRD curve of the FATIMA plus the electronics set-up that was calibrated using γ-γ cascades from a standard ^{152}Eu γ-ray source and the in-beam reaction $^{48}\mathrm{Ti}(\mathrm{n},\gamma)^{49}\mathrm{Ti}$ [13]. Some 82% of the data are nearly free of background due to the use of an additional EXILL (Ge) gate to select a triple γ-ray cascade. For the EXILL&FATIMA experiments, a PRD uncertainty of 10 ps has been achieved which includes possible systematic errors [13].

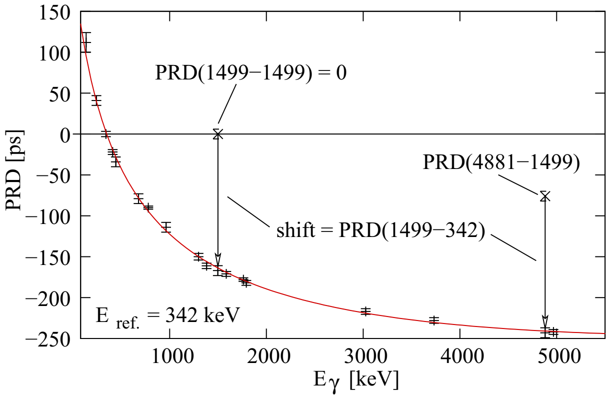

Figure 3. The (mean) PRD curve of the FATIMA plus the electronics set-up of the EXILL&FATIMA spectrometers, adjusted for the refence energy of 342 keV. The PRD-calibration procedure is to adjust the two data points of prompt γ-γ cascades by parallel shifts to fit a single smooth PRD curve. Referring to the decay transition of a cascade, the measured $\mathrm{PRD}(\Delta E_\gamma)$ is plotted at the energy of the feeding γ ray, while the PRD at the decay energy vanishes according to Eqs. (9) and (10) and thus provides the second data point with same statistical uncertainty.

The major systematic error that can be induced in GCD analysis is due to the Compton background underneath the full-energy peak of interest which contributes to the FATIMA time spectra. Fig. 4 illustrates a test case on the lifetime determination of the 2_1^+ state in ^{100}Zr. The results are from triple Ge-LaBr-LaBr (or Ge-LaBr-Ge) coincidences out of 8 TB of data digitally acquired triggerless from a 12 days measurement on cold-neutron-induced fission fragments of ^{235}U. As shown in Fig. 4a, a relatively large Compton background is obtained due to high γ-ray multiplicity of over 100 nuclei produced in such experiments. However, the double-gated coincidence spectra are clean, meaning that no other γ rays are observed in the vicinity of the peak at 352 keV, that could falsify the lifetime determination. The delayed and anti-delayed FATIMA time spectra of the 352-212-keV cascade shown in Fig. 4b are nearly mirror symmetric which indicates the high quality of the FATIMA plus electronics set-up. By assuming the γ-Compton (full-energy peak vs. Compton) time response to be nearly prompt, the lifetime can be extraced directly from the slope of the time spectra [1].

The GCD method is sensitive to Compton background contributions to the time spectra. As illustrated in Fig. 4c, the 212-keV γ vs. 352-keV Compton time response can be derived from a set of time spectra generated by setting gates in the Compton background around and above 352 keV. Taking into account the peak-to-background ratio $\Pi = 2.6(2)$ of the 352-keV peak in the FATIMA LaBr_3 coincidence spectrum shown in Fig. 4a, the net γ-γ centroid difference ΔC_{net} is derived from the time shift [here +649(20) ps] between the measured centroid difference and the γ-Compton time response $\Delta C_{\mathrm{Compton}}$ at 352 keV using [3, 13]:

$$\Delta C_{\mathrm{net}} = \Delta C + \frac{\Delta C - \Delta C_{\mathrm{Compton}}}{\Pi}. \qquad (12)$$

By inserting the net centroid difference in Eq. (11) with $\mathrm{PRD}(352) = -61(10)$ ps, the mean lifetime of the first 2^+ state in ^{100}Zr of $\tau(2_1^+) = 821(12)$ ps follows, which is in good agreement with the results obtained using the slope method and also confirms the experimental weighted average of $\tau = 851(43)$ ps reported in Ref. [15]. Further tests and new measurements on short lifetimes down to the limit of about 10 ps also confirmed the method [13, 16–18].

4 Conclusions

At the PF1B collimated cold-neutron-beam line of the Institut Laue-Langevin, Ge-gated γ-γ fast-timing measurements of excited states have been performed on around 80 nuclei from neutron-capture and neutron-induced fission experiments using 16 $\mathrm{LaBr}_3(\mathrm{Ce})$ detectors. The complex data sets are analyzed using the newly developed GCD method which provides a simple algorithm and thus a rapid tool to generate the fast-timing-array time spectra. The mirror-symmetric GCD method is picosecond sensitive making it possible to derive a precise time correction related to the Compton background underneath the full-energy peaks of the γ-γ cascades. First high-precision results of lifetimes down to the limit of about 10 ps have been reported. The GCD method is a powerful tool which allows to test the set-up of a fast-timing array. Deviations from smooth γ-γ time walk (the PRD curve) are quickly identified and indicate systematic shifts, e.g. induced by cross-talk events.

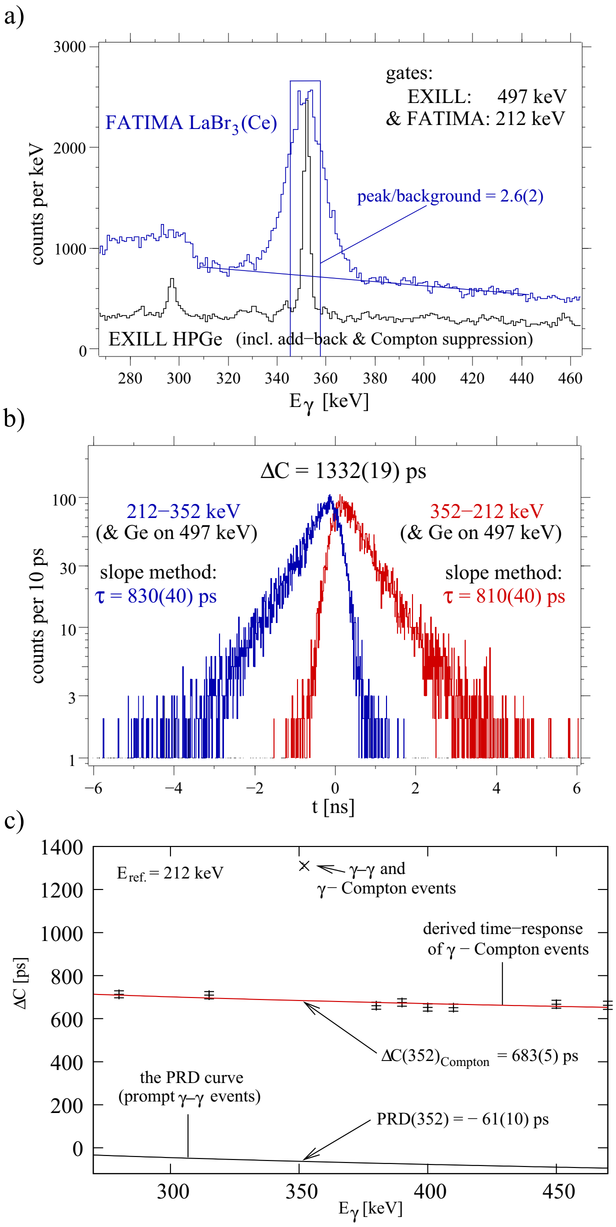

Figure 4. a: Comparison of the double-gated (Ge+LaBr) FATIMA and EXILL coincidence spectra, showing the cleanliness of the data in the region of the 352-keV peak. b: The two EXILL-gated FATIMA time spectra of the 352-212-keV γ-γ cascade in ^{100}Zr. According to the peak-to-background ratio, about 40% of the data are due to γ-Compton events. c: Illustration of the γ-Compton time-correction procedure by the centroid difference diagram for the reference energy of 212 keV (common gate of all time spectra). The PRD curve is adjusted by a parallel shift to cross the energy axis at the reference energy.

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