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Precision measurement of sub-nanosecond lifetimes of excited nuclear states using fast-timing coincidences with $LaBr_3(Ce)$ detectors

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HIGHLIGHTS

- This paper reports on new detection systems for identification of gamma decays.
- It focusses on using arrays of halide-scintillation detectors in coincidence mode.
- These can determine lifetimes of nuclear states at the sub-ns level.
- Plans for new LaBr3(Ce) arrays for studies of exotic nuclei are presented.

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ABSTRACT

Precision measurements of electromagnetic (EM) transition rates enable tests of models of internal nuclear structure. Measurements of transition rates can be used to infer the spin and parity differences between the initial and final discrete nuclear excited states via which the EM transition takes place. This short conference paper reports on developments of detection systems for the identification of discrete energy gamma-ray decays using arrays of halide-scintillation detectors acting in coincidence mode, which can be used to determine electromagnetic transition rates between excited nuclear states in the sub-nanosecond temporal regime. Ongoing development of a new multi-detector $LaBr_3(Ce)$ array for studies of exotic nuclei produced at the upcoming Facility for Anti-Proton and Ion Research (FAIR) as part of the NUSTAR–DESPEC project are presented, together with initial results from pre-NUSTAR implementations of this array for nuclear structure studies of neutron-rich fission fragment radionuclides at ILL-Grenoble, France and RIBF at RIKEN, Japan.

1. Introduction

1.1. Measurements of decay rates from nuclear excited states

Electromagnetic (EM) transition rates between excited states of atomic nuclei can provide direct information on the underlying structural makeup and nucleonic configurations of the nucleus. Decay lifetimes of excited nuclear states range from femtoseconds (Schwarzschild and Warburton., 1968; Nolan and Sharpey-Schafer, 1979) to more than 10^{15} years (Cumming and Alburger, 1985) depending on the energy of the transitions and the spins and parities of the initial and final nuclear states.

The EM rate, which corresponds to the probability of decay per unit time, can be written as (Bohr and Mottelson, 1999).

$$T_{fi} = \frac{1}{\tau} = \frac{8\pi(L+1)}{\hbar((2L+1)!!)^2}\left(\frac{E_\gamma}{\hbar c}\right)^{2L+1} B\left(\lambda L;\ I_i \rightarrow I_f\right)$$

The measurement of the mean lifetime of decay for an electromagnetic transition of a defined decay energy (i.e. a discrete gamma-ray energy transition) gives direct information on the reduced matrix element linking the initial and final quantum states via that particular EM multipole operator. A range of measurement techniques can be applied to the determination of accurate lifetime evaluations for excited nuclear states (Schwarzschild and Warburton., 1968; Nolan and Sharpey-Schafer, 1979) depending on the time regime involved. For transitions in the tens of nanosecond regime and higher, the (fast) electronic timing method has been well established (Mach et al., 1989, 2005)

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2. $LaBr_3$(CE) detector dimensions, characteristics and performance for use in current multi-detector arrays

The idealized gamma-ray spectrometric detectors would have a combination of excellent full-energy peak resolution, together with good full-energy peak detection efficiency and very fast, sub-nanosecond timing characteristics. The energy resolution associated with multi-detector arrays of hyper-pure germanium detectors (Mach and Fraille, 2014; Eberth and Simpson, 2008; Beausang and Simpson, 1996; Lee et al., 2003) and the timing capabilities for gamma-ray detection from BaF_2 scintillation detectors (Mach et al., 2005; Laval et al., 1983; White et al., 2007) represent the current precision limits for standard nuclear spectroscopic measurements.

Developments in the production and operation of cerium-doped lanthanum tri-bromide ($LaBr_3$(Ce)) scintillators has resulted in a gamma-ray detection material with an acceptable energy resolution combined with fast-timing characteristics. Early examples of measurements using $LaBr_3$(Ce) detector systems for transition rate measurements between discrete states include radioactive decay studies of neutron-rich radionuclides species by Mach and collaborators following using the Advanced Time-Delayed $\beta-\gamma-\gamma$ (t) method for fast beta-gamma coincidence measurements (Smith et al., 2008). Other examples of in-beam spectroscopic measurements of nuclear excited-state lifetimes have since been reported in systems produced via light-ion induced fusion-evaporation and light-ion transfer reactions, in particular a following the ongoing development of the mixed, combined HpGe-$LaBr_3$ spectrometer array ROSPHERE, based at the IFIN-HH laboratory, Bucharest (Marginean et al., 2010; Kisyov et al., 2011; Mason et al., 2012, 2013; Alharbi et al., 2013, 2014; Alharbi, 2012; Nita et al., 2014), including measurements of mean lifetimes below 40 ps (Bocchi et al., 2014).

An array of $LaBr_3$(Ce) detectors, ultimately for use with as part of the DEcay SPECtroscopy (DESPEC) collaboration within the NUSTAR collaboration at the future Facility for Anti-Proton Ion Research (FAIR) facility is currently under development (Regan, 2012; Roberts et al., 2014; Podolyak, 2014) with an initial design of 32 detectors surrounding the final focal plane of the Super Fragment Separator at FAIR. For this initial design, 32 cylindrical $LaBr_3$(Ce) detectors, each of length 2″ and diameter 1.5″ and coupled to a Hamamatsu R9779 fast-timing photo-multiplier tube, have been purchased via the UK Science and Technology Facilities Council (STFC) NUSTAR grant. These detectors have been characterized using a range of source measurements at the Environmental Radioactivity Laboratory at the University of Surrey, with energy resolutions and timing FWHM down to 210 ps measured for gamma-ray energies of 1332 keV using a ^{60}Co coincidence source (Roberts et al., 2014) and typical full energy peak resolutions of ~3% for 662 keV (Regan, 2012; Roberts et al., 2014). Fig. 1 shows a photograph of the support frame structure and a number of $LaBr_3$ detectors in-situ which has been constructed as part of this project.

Fig. 1. Photographs of the support frame for the decay spectroscopy FAst-TIMing Array (FATIMA) of LaBr3 detectors for future use within the DESPEC project at FAIR (Regan, 2012).

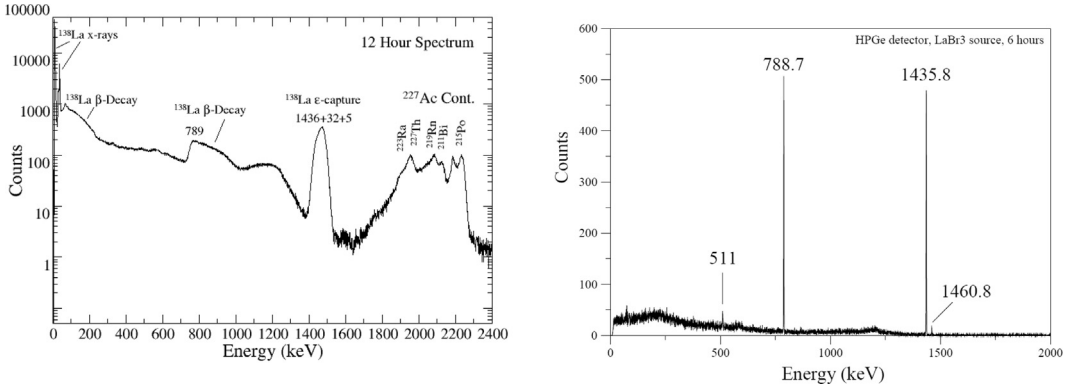

Fig. 2. (Left) $LaBr_3$ detector spectrum from raw detector placed in passive graded lead shield showing the internal radioactivity from the ^{138}La decays. Note the presence of signals arising from both ^{138}La decay and members of the ^{220}Ac decay chain. (Right) Emitted gamma ray spectrum from a $2'' \times 1.5''$ diameter $LaBr_3(Ce)$ detector measured in a passively shielded HpGe detector for a period of 6 hours.

The $LaBr_3(Ce)$ detectors also exhibit spectral features associated with the internal radioactivity from the decay of ^{138}La which is present in the lanthanum detector material. This primordial radionuclide makes up approximately 0.09% of naturally occurring lanthanum and has a radioactive half-life of 1.0×10^{11} years, decaying by both electron capture (65.6%) and β^- decay (34.4%) to the lowest-lying spin/parity 2^+ states in ^{138}Ba (at 788.7 keV) and ^{138}Ce (1435.8) respectively (Sonzogni, 2003). The detector also contains internal radioactivity associated with the natural decay chain of ^{227}Ac, which has a 22 year half-life and occurs as a member of the ^{235}U (actinium) $4n+1$ natural decay series (e.g., (Malain et al., 2012)). Chemically speaking, actinium is similar to lanthanum and is present in trace amounts in all $LaBr_3(Ce)$ detectors. ^{227}Ac decays to stable ^{207}Pb through a cascade of discrete alpha decays which can be identified as an internal radioactive source within the final detector response. The observed alpha-particle energies from these decays are quenched in energy compared with direct measurements of external gamma-rays of the same energy. Fig. 2 shows the effect of the internal radioactivity from the ^{138}La and the^{227}Ac decay series on the internal response of the $LaBr_3(Ce)$ detector itself. Also shown for reference is the measured gamma-ray emissions from one of the $LaBr_3(Ce)$ detectors in a passively shielded HpGe detection system within the Environmental Radiation Laboratories at the University of Surrey (Malain et al., 2012). As for signals measured which arise from the $LaBr_3(Ce)$ internal radioactivity sources in external detectors, only the discrete gamma rays from the decay of the excited states populated in the ^{138}La are observed. The internal radioactivity associated with such detectors is of the order of $\sim$1 Bq/cm^3. While this gives rise to an easily measureable background signal which can be used as internal calibration standard for the detector energy response, in the presence of stringent coincidence conditions with other correlated signals from radioactive decay and/or gamma rays which occur within 10 ns or so of the signal of interest, this level of background activity does not affect most spectroscopic studies.

The timing responses associated with spectroscopy grade $LaBr_3(Ce)$ detectors used in coincidence mode has been established down to the $\sim$10 ps level by Regis and collaborators (Régis et al., 2012, 2010) using the mirror symmetric centroid shift method. Off-line software corrections to event-by-event gamma-ray coincidence data, for the full-energy-peak dependent, prompt-timing response for each detector are applied using empirical information from a well-understood prompt coincident decay source, such as ^{152}Eu. Regis and collaborators have demonstrated that, that if sufficient coincident statistics can be measured from a point-like, localized source, discrete level lifetimes at the tens of picoseconds levels are measureable using $LaBr_3(Ce)$ coincident fast-timing.

3. Examples of current nuclear spectroscopy gamma-ray arrays incorporating $LaBr_3(Ce)$ detectors

Published lifetime results from a number of experiments have been reported using a coincidence array consisting of $LaBr_3(Ce)$ and HpGe detectors at the IFIN-HH laboratory in Bucharest, Romania for measurements of nuclear excited state lifetimes in the sub-nanosecond time range. The current evolution of this array, known as ROSPHERE (Marginean et al., 2010), has allowed a wide range of nuclear structure questions to be investigated by the establishment of precision, sub-nanosecond lifetime measurements. These include studies of magnetic quadrupole (M2) single particle transitions approaching the $N=20$ magic number (Mason et al., 2012; Alharbi, 2012); measurements of transitions rates between competing shell model configurations in the $N=80$, two-neutron-hole system ^{136}Ba (Alharbi et al., 2013); Low-lying electric dipole (E1) transition probabilities in the transitional nucleus ^{136}Ce (Alharbi et al., 2014); measurements of the lifetime and the related inference of the magnitude of the nuclear quadrupole deformation for the first excited state in the neutron-rich shape-transitional system ^{188}W (Mason et al., 2013); and studies of nuclear structure in neutron-rich Cu isotopes (Nita et al., 2014; Bocchi et al., 2014). Fig. 3 shows example spectra from the HpGe and $LaBr_3(Ce)$ detectors from the ROSPHERE array in the discrete-line study of ^{136}Ce by Alharbi et al. (2014) populated in a heavy-ion fusion-evaporation reaction at the IFIN-HH facility in Bucharest, Romania. Note the clarity of the $LaBr_3(Ce)$ spectra identifying decays from the yrast cascade in this nucleus when the coincidence requirement of observing another discrete transition in the detectors is applied.

Looking towards the future, the thirty-two $1.5''$ diameter and $2''$ length LaBr3(CE) detectors which make up the UK fast-timing project, within the FATIMA collaboration have already been incorporated into a number of other combined HpGe-LaBr3(CE) detection systems and used in a range of nuclear structure studies. eighteen of these LaBr3(CE) detectors have been used in conjunction with former RISING stopped beam germanium detector array for both isomer and beta-delayed gamma-ray spectroscopy measurements. Fig. 4 shows a CAD design drawing of a section of this combined array. Known as EURICA, this instrument consists of twelve, seven-element germanium cluster detectors and Is positioned at the focal plane of the big RIPS fragment Separator at the RIKEN laboratory, Japan (Regan et al., 2013; Söderström et al., 2013; Browne et al., 2014). EURICA has been used to measure a range of neutron-rich nuclei following production via projectile fission. for example, fast-timing coincidences between the LaBr3(CE) detectors in this array and a fast β-particle decay signal at the focal plane have allowed the determination of the lifetimes (and associated transition quadrupole moments, in a range of neutron-rich

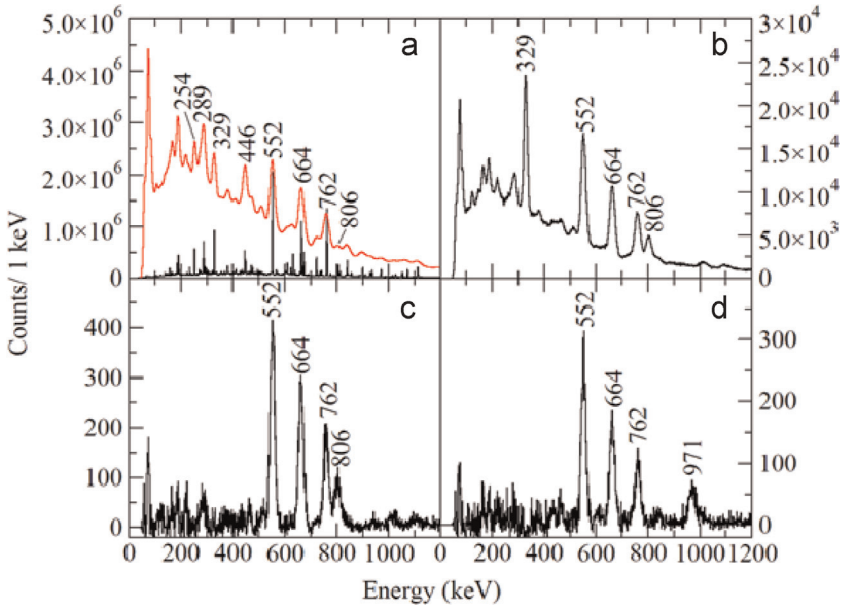

Fig. 3. An example of the use of discrete-line coincidence gating to select specific decay cascades populated in nuclear reactions, using the ROSPHERE mixed HpGe-LaBr3(Ce) gamma-ray spectrometer at IFIN-HH Bucharest. (a) Total projection for all HpGe and $LaBr_3$(Ce) detectors from the reaction $^{124}Sn+^{16}O$ at a beam energy of 68 MeV (Alharbi et al., 2014) used in the study of yrast states in ^{136}Ce with the ROSPHERE gamma-ray array. (b) total projection of all $LaBr_3$(Ce) detectors with a coincidence gate on 971 keV transitions in the HpGe detectors; (c) as (b) but with an additional condition of 329 keV gate on the $LaBr_3$(Ce)_detectors; (d) as (b) but with an additional coincidence condition on the 806 keV transition in the $LaBr_3$(Ce) detectors.

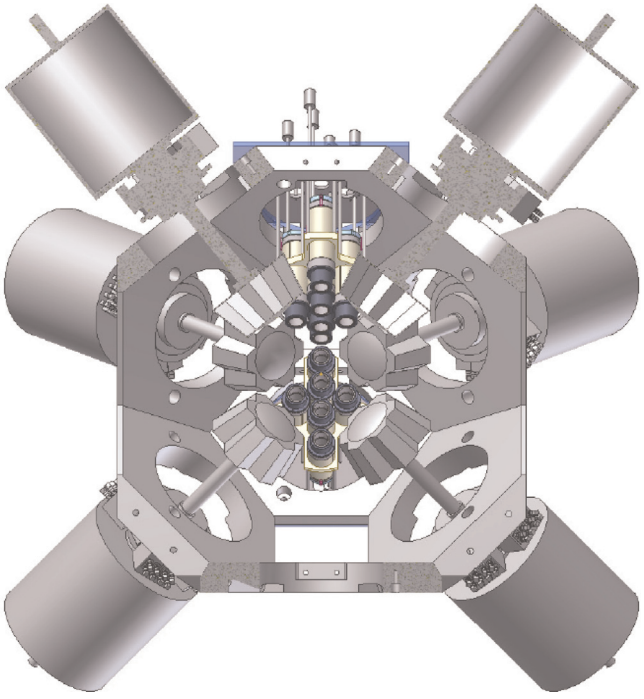

Fig. 4. CAD drawing of partial EURICA gamma-ray array (Söderström et al., 2013) showing 12 of the 18 $LaBr_3$(Ce) detectors used in its configuration.

zirconium isotopes following the β-decay of their yttrium parent nuclei (Browne et al., 2014).

3.1. EXILL with FATIMA at ILL-Grenoble

Sixteen $LaBr_3$ detectors (8 from the STFC funded groups and 8 more from the IFK Koln and TU Darmstadt groups) were used together with the EXOGAM HpGe Clover detectors for experiments using thermal neutrons at the ILL-Grenoble facility in 2013, see Fig. 5. These experiments included in-beam spectroscopy of

Fig. 5. Photograph of the target position of the EXILL+FATIMA set-up at ILL-Grenoble. This array combined the clover HpGe detectors in the EXOGAM array with 16 $LaBr_3$ detectors for fast-timing measurements of prompt fission fragments.

prompt fission fragments produced following cold-neutron induced fission on a ^{235}U target placed in the center of the array including ^{235}U(n,f) (Blanc et al., 2013; Régis et al., 2014). The analysis of these data included, in general quadruple, hyper-cube type energy coincidences (i.e., 2 discrete line gates on HpGe signals for isotope and cascade identification, and 2 further coincidences on $LaBr_3$(Ce) signals for identified discrete line transitions which bootstrap the energy level for which the lifetime/transition rate is to be measured.

4. Summary and conclusions

Detector arrays for nuclear spectroscopic measurement which include the capability for coincidence spectroscopy between discrete gamma-ray transitions using halide scintillation detectors are rapidly becoming mainstream tools in nuclear structure

physics research worldwide. The ability of $LaBr_3$(Ce) detectors to give suitably fast timing information coupled with acceptable energy resolution makes them an ideal tool for discrete nuclear spectroscopy studies. This is particularly true in cases where (a) the spectral level density is not too high, for example following tagged beta-decays of exotic nuclei produced at radioactive ion beam facilities and/or (b) when used in coincidence with a complementary array of hyper-pure germanium detectors which can be used as a channel/decay path selection device to isolate the particular scintillator coincidences across a nuclear level of particular interest. Other related, (more) 'background free' detector materials, such as $CeBr_3$ are also of interest in this area of research (Fraile et al., 2013) which can also be transitioned from fundamental research studies, to practical measurements of discrete radionuclide releases for health physics and nuclear power/waste management purposes.

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