

Applications of LaBr₃(Ce) Gamma-ray Spectrometer Arrays for Nuclear Spectroscopy and Radionuclide Assay

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Abstract. An overview of the use of discrete energy gamma-ray detectors based on cerium-doped LaBr₃ scintillators for use in nuclear spectroscopy is presented. This review includes recent applications of such detectors in mixed, 'hybrid' gamma-ray coincidence detection arrays such ROSPHERE at IFIN-HH, Bucharest; EXILL+FATIMA at ILL Grenoble, France; GAMMASPHERE+FATIMA at Argonne National Laboratory, USA; FATIMA + EURICA, at RIKEN, Japan; and the National Nuclear Array (NANA) at the UK's National Physical Laboratory. This conference paper highlights the capabilities and limitations of using these sub-nanosecond 'fast-timing', medium-resolution gamma-ray detectors for both nuclear structure research and radionuclide standardisation. Potential future application of such coincidence scintillator arrays in measurements of civilian nuclear fuel waste evaluation and assay is demonstrated using coincidence spectroscopy of a mixed ^{134,7}Cs source.

1. Introduction: The Use of LaBr₃(Ce) Detectors for Nuclear Gamma-ray Measurements.

The science of nuclear spectrometry requires careful measurement of the characteristic decay properties of a range of radionuclides. These can have applications in nuclear waste disposal, radiopharmaceutical standardisations and also in fundamental studies of the internal structure of atomic nuclei. The characteristic, discrete-energy gamma-ray emissions from excited states populated following radioactive decay provide unique signatures which can be used both for identification and quantification of different radionuclides species in mixed-material sources. The workhorse detectors for such studies over the last three decades have been hyper-pure germanium (HPGe) semiconductor detectors which have excellent gamma-ray energy resolution (<2 keV FWHM at 1.33 MeV full energy peak) but have to be operated at liquid nitrogen temperature in order to overcome thermal excitation noise effects.

Over the course of the last decade there has been a significant development in nuclear instrumentation based on detectors made from halide scintillation materials for use in gamma-ray spectroscopy and spectrometry. In particular, the inorganic scintillator Lanthanum Tribromide with Cerium doping (LaBr₃(Ce)) is becoming a mainstream tool in nuclear spectroscopy research



experiments [2-20]. This material has a significantly poorer energy resolution compared to HPGe (~40 keV FWHM at 1.33 MeV [7]) but is much better than other scintillator materials for gamma-ray measurement such as NaI(Tl) or CsI [1]. It also has some operational advantages compared to HPGe in that its intrinsic timing resolution is sub-nanosecond across a wide energy range (from ~100 keV up to more than 4 MeV) reduces random events in energy coincidence analyses where two or more discrete energy gamma rays are emitted in a sequential cascade following a radioactive decay. It can also provide precision determinations of electromagnetic decay lifetimes of intrinsic, excited nuclear states, thereby providing tests of physical models of nuclear structure.

This ANSRI conference paper reviews the construction and application of a range of gamma-ray spectrometer arrays worldwide which utilise contemporary $\text{LaBr}_3(\text{Ce})$ detector technology and provide examples of analysis for nuclear waste assay and nuclear structure physics measurements.

2. The National Nuclear Array – NANA

The National Nuclear Array (NANA) has been commissioned at the UK National Physical Laboratory [2-4]. This array comprises 12 $\text{LaBr}_3(\text{Ce})$ detectors, each of which is a cylinder of length 51 mm and circumference 38 mm. The current array design consists of a central ring of 8 symmetrically-spaced $\text{LaBr}_3(\text{Ce})$ detector modules with an additional ring of 4 detectors off-set at backward angles. These are the same detector modules and geometries as used in the DESPEC-FATIMA gamma-ray spectrometer developed for use in nuclear structure decay experiments at Radioactive Ion Beam research facilities such as at GSI/FAIR and RIBF-RIKEN [5-8].

Figure 1 shows photographs of the NANA in-situ at the National Physical Laboratory. The $\text{LaBr}_3(\text{Ce})$ detectors are coupled to Hamamatsu R9779 photomultiplier tubes (PMTs) were are chosen to maximise the timing capabilities of these detectors. The energy signals from the PMTs are taken directly into CAEN V1751, 1 GHz digitisers which provide independent but synchronised time-stamped, energy signals for each input channel. The digitised nanosecond timing allows event-by-event coincidence information between gamma rays emitted from a discrete cascade to be temporally correlated; in turn, logical gating conditions can be set on particular gamma-ray energies which define a unique energy-level decay sequence characteristic for a particular radionuclide species. This coincidence gating property can be applied event-by-event in off-line software data analysis and allows $\text{LaBr}_3(\text{Ce})$ -based detection arrays such as NANA the potential to identify relatively low-activity concentrations. This can be particularly useful when analysing mixed-waste source matrices which may emit a large number of different energy gamma rays resulting in what, at first instance, leads to a rather messy and hard to interpret ‘singles’ spectrum.



Figure 1 The NANA gamma-ray spectrometer at the UK National Physical Laboratory, comprising twelve individual $\text{LaBr}_3(\text{Ce})$ gamma-ray detector modules.

2.1. Example of use of NANA for Radionuclide Assay: $^{134}\text{Cs}/^{137}\text{Cs}$ Identification and Separation.

The NANA allows the use of coincidence gamma-ray spectroscopy in the evaluation of radionuclide materials. As an example, we have taken data using six independent modules within the NANA array from gamma rays emitted from a well characterised mixed $^{134}\text{Cs}/^{137}\text{Cs}$ source. Both of these radioisotopes would be expected to be present in nuclear waste fuel; (a) ^{137}Cs , which has a half-life of 30 years [22], as a direct fission fragment daughter and (b) ^{134}Cs ($T_{1/2}=2.0652$ years [23-24]) which is formed in-situ in nuclear reactor fuels following thermal neutron-capture on the ^{133}Cs fission daughter residue which builds up during normal reactor operation.

The gamma-ray decays following the radioactive decay for these two isotopes are notably different with ^{137}Cs decaying primarily to a single excited, spin/parity $I^\pi=11/2^-$ metastable state ($T_{1/2}=2.55$ mins) which then emits a single 662 keV gamma-ray to the $I^\pi=3/2^+$ ground state of the ^{137}Ba daughter radionuclide. By contrast, the spin/parity $I^\pi=4^-$ ground state of ^{134}Cs decays both by electron capture to ^{134}Xe and also by β^- decay to excited states in ^{134}Ba [23-24]. The left hand side of Figure 2 shows a simplified partial level schemes for the states populated in ^{134}Ba following the radioactive decay of ^{134}Cs . Note that there is a range of multi-gamma-ray cascades which feed the 605 keV yrast $I^\pi=2^+$ state in the ^{134}Ba daughter. The right hand side of Figure 2 shows the summed singles spectra for 6 detectors of the NANA array from the mixed $^{134,7}\text{Cs}$ source. The raw, energy-calibrated spectra show the well-understood signatures of arising from the internal activity within the $\text{LaBr}_3(\text{Ce})$ detectors from (i) the decay of the ^{138}La primordial isotope [25] and (ii) a cascade of alpha emitters from the members of the actinium natural decay series ($4n+3$) chain headed by ^{227}Ac which are intrinsic to the detector material. A normalized fraction of the ‘sourceless’ spectrum (i.e., a spectrum taken from detector signals measured with no source present in the NANA source position) has been subtracted and this clearly shows and is able separate discrete lines from the decay of ^{137}Cs (662 keV) and ^{134}Cs (569, 605, 796, 1038, 1168 and 1365 keV).

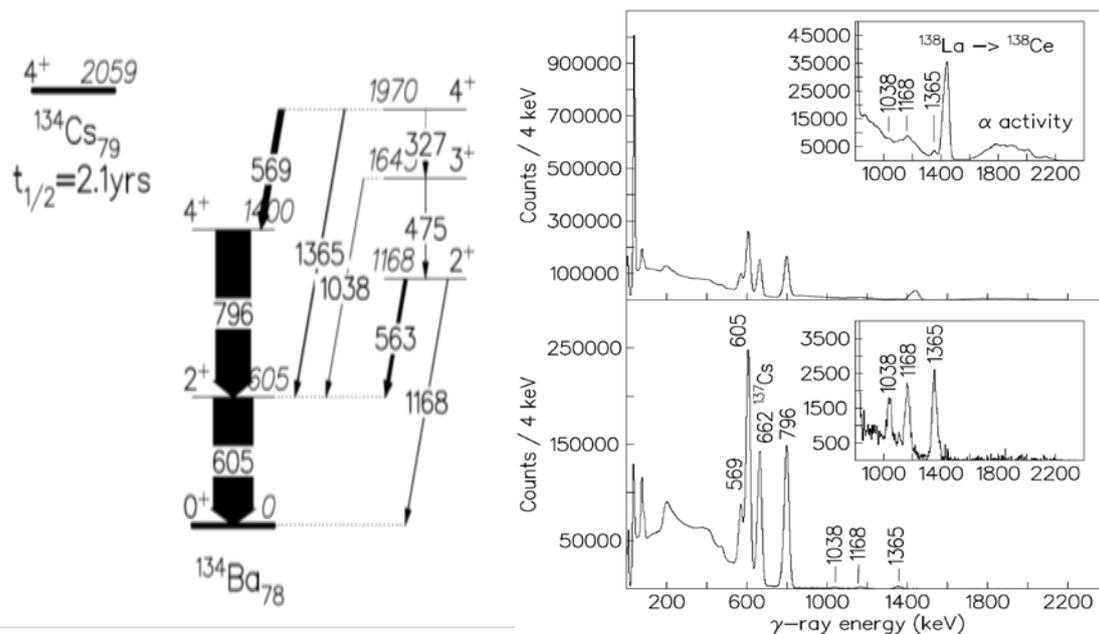


Figure 2 (Left) Partial decay scheme of states populated in ^{134}Ba following the radioactive decay of the nuclear waste product ^{134}Cs [23,24]. (Right) summed singles (upper panel) and background-subtracted (lower panel) $\text{LaBr}_3(\text{Ce})$ spectra using NANA for a mixed $^{134,137}\text{Cs}$ source. Note the clear identification of the weak transitions above 1 MeV in the background subtracted spectrum.

The selection power of NANA in using the coincidence method to isolate gamma-ray decay cascades associated with particular radionuclide decays is demonstrated in Figure 3. These are the same data as shown in Figure 2 but with an offline software-sorting condition applied that at an energy signal should be registered in at least two separate detectors of the NANA array within a time window of ± 5 ns. For these data, six of the full complement twelve NANA detector modules were operational. The left hand side of Figure 3 shows the two-dimensional gamma-ray-energy coincidence matrix for these data. The diagonal, negative gradient stripes in this matrix correspond to Compton scattered events where the sum of energies in the two coincident detectors sum to a single (Compton scattered) discrete energy. Notable lines of this nature include the 1435.8 keV transition emitted from the internal ^{138}La activity in the detector material [25]. By contrast, the main two-dimensional peaks within this energy matrix correspond to the full-energy peak coincidence events from the decay of ^{134}Cs into excited states in ^{134}Ba , for example the coincidence pairs at {605,796} and {605;563/569} [23,24]. The right hand side of Figure 3 shows a total projection (i.e. sum) of all coincidence with ≥ 2 independent $\text{LaBr}_3(\text{Ce})$ events measured within a ± 5 ns coincidence window. Note that there is no lines present in this spectrum associated with the 662 keV transition from the ^{137}Cs decay, even though this line is clearly observed in the singles spectra for the same data shown on the left hand-side of Figure 2. Furthermore, when off-line coincidence conditions are set whereby a discrete full-energy peak is measured in one of the two coincidence detectors, the projections can be used to demonstrate and isolate different decay paths within the ^{134}Ba nucleus. For example, when a coincidence is required with the 1365 keV transition, only the 605 keV $2^+ \rightarrow 0^+$ transition from the decay of first excited state in ^{134}Ba is evident in the spectrum, with the 796 keV and other transitions absent.

The use of $\text{LaBr}_3(\text{Ce})$ detectors has been mooted recently for application in nuclear waste fuel assay [26]; the additional parameter dimension which is allowed by discrete gamma-ray coincidence analysis could allow an improvement of this technique for more detailed assay of more weakly populated fission residues if their decay incorporates a coincidence decay sequence such as the ^{134}Cs example shown in the current work.

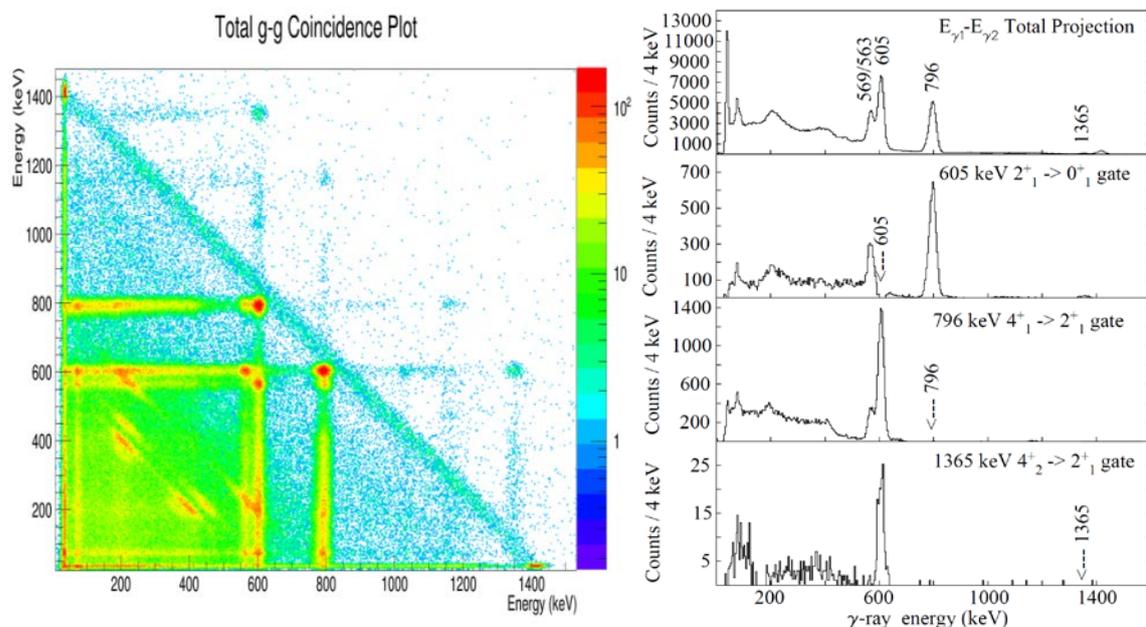


Figure 3 (Left) gamma-ray energy coincidence matrix for the mixed $^{134,137}\text{Cs}$ source shown in Figure 2 using 6 individual detectors in the NANA array; (Right) Total Projection (upper spectrum) and Compton-background subtracted gates on the matrix showing coincidence transition in ^{134}Ba populated following the decay of ^{134}Cs .

3. Hybrid-LaBr₃(Ce)-HPGe Gamma-ray Coincidence Arrays in Nuclear Structure Research:

3.1. The DESPEC-FATIMA Array for FAIR and Related sub-arrays.

A number of coincidence gamma-ray spectrometer arrays have been constructed and commissioned worldwide to exploit the timing characteristics of LaBr₃(Ce) detectors in nuclear spectroscopy with focus on the measurement of absolute electromagnetic transition rates associated with decays from excited, particle bound nuclear states. The detectors in the NANA are also used in the DESPEC Fast-Timing Array (FATIMA). This array consists of three concentric rings of twelve LaBr₃(Ce) detectors (36 detectors in total) and has been designed for gamma-ray spectroscopic measurement following decays in nuclei with very exotic proton-to-neutron ratios produced at high-energy nuclear projectile fragmentation facilities such as GSI/FAIR and RIBF-RIKEN [5-8].

Some of these LaBr₃(Ce) detector modules have been incorporated into mixed, 'hybrid' arrays which combine HPGe detectors for high-resolution gamma-ray selection and LaBr₃(Ce) modules for coincident nuclear fast-timing measurements. An example is the EXILL+FATIMA set-up which was used to measure prompt gamma-ray coincidences following the cold-neutron-induced, prompt fission of ²³⁵U at ILL Grenoble [11-12]. EXILL+FATIMA comprised eight LaBr₃(Ce) detectors used in concert with eight, four-element HPGe Clover detectors in a compact geometry and was used to study the internal structure of neutron-rich, prompt-fission residues. The system typically used quadruple coincidence events (2 x HPGe and 2 x LaBr₃(Ce)) to isolate individual fission fragment cascades from the hundreds of identified prompt species populated in this reaction mechanism. The EXILL-FATIMA set-up has been used to measure nanosecond excited state lifetimes for the excited states of neutron-rich nuclei such as ¹⁰⁰Zr [11] and ⁹⁰Kr [12].

The DESPEC-FATIMA detectors have also been incorporated at the Radioactive Ion Beam Facility (RIBF) at RIKEN, Japan for related nuclear structure studies of some the most neutron-rich nuclear species identified to date. A mixed, hybrid array of 18 DESPEC-FATIMA LaBr₃(Ce) detectors were coupled to twelve, seven-element HPGe 'CLUSTER' detectors at the focal plane of the BigRIPS projectile separator at RIKEN. The main application for this array was gated isomer and beta-delayed nuclear spectroscopy in very neutron-rich nuclei which were produced following the high-energy projectile fission of a ²³⁸U primary beam [13]. Recent physics highlights using this array include the measurement of the first excited state lifetimes in ^{104,6}Zr which are 8 and 10 neutron heavier respectively than the most neutron-rich stable Zirconium isotope [14]. This new experimental information can be used to determine the low-lying shape of these mesoscopic physical systems and allows a comparison with state of the art nuclear mean-field energy density functional calculations which predict the evolution of nuclear structure in such nuclei of astrophysical interest. Twenty-five of the DESPEC-FATIMA LaBr₃(Ce) detectors were coupled together with 55 HPGe Compton suppressed detectors of the GAMMASPHERE array at Argonne National Laboratory in late 2015 for an experimental measurement campaign of prompt and delayed decays from spontaneous fission residues from a sealed ²⁵²Cf source [8]. An array of 8 LaBr₃(Ce) FATIMA detectors has also been deployed at the focal plane of the RITU gas-filled mass separator at JYFL- Jyvaskyla, Finland for lifetime and nuclear deformation measurements in N~74 isotones following isomeric decay [27].

Experimental plans for the 2016-17 period include combining these FATIMA detector modules with the AGATA gamma-ray tracking spectrometer for a nuclear structure campaign at the GANIL laboratory in Caen, France for studies of neutron-rich nuclear systems using heavy-ion induced deep-inelastic reactions and also with 24 clover HPGe detectors to form the NuBALL hybrid gamma-ray spectrometer to be used at IPN-Orsay, Paris France for fast-neutron induced fission measurements.

3.2. RoSPHERE

The first major hybrid $\text{LaBr}_3(\text{Ce})$ -HPGe array to be used in continuous mode is the **RO**manian array for **SP**ectroscopy in **HE**avy ion **RE**actions (ROSPHERE) at IFIN-HH, Magurele, Romania [28,29]. Its most recent configuration comprises 25 individual detectors, fourteen 50-60% relative efficiency Compton Suppressed HPGe spectrometers and 11 $\text{LaBr}_3(\text{Ce})$ modules, with five detectors each set into five concentric rings at angles of 37° , 70° , 90° , 110° and 143° to the beam direction. Figure 4 shows photographs of the ROSPHERE array with the open vacuum target chamber.

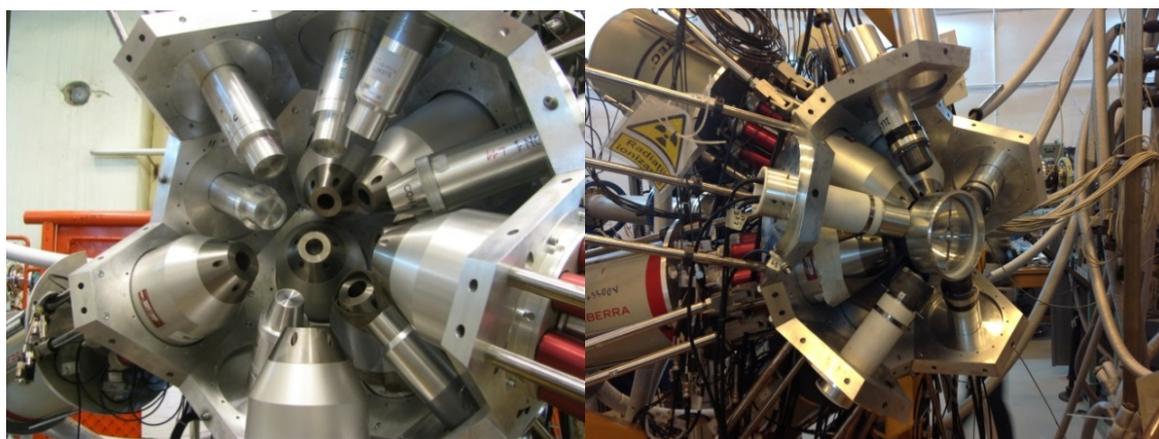


Figure 4 Insert of part of the ROSPHERE hybrid gamma-ray at IFIN-HH, Romania consisting of Compton Suppressed HPGe detectors and $\text{LaBr}_3(\text{Ce})$ for fast-timing measurements [29].

ROSPHERE has been used to study a wide range of nuclear structure phenomena in nuclear created following heavy-ion reactions from beams produced by the 9 MV tandem van de Graaff accelerator at IFIN-HH. Such studies have included evaluation of single-particle transitions around the $N=20$ [15], $N=82$ [16], and $N=126$ [17] magic numbers; measurements of low-lying octupole ($E3$) excitations in neutron-rich copper nuclei [18,19]; shape evolution from prolate to oblate deformations in tungsten isotopes [20]; and the robustness of the K -quantum number associated with axial symmetry in prolate-deformed Cerium isotopes [21].

Figure 5 shows example spectra from ROSPHERE following an 80 MeV ^{18}O beam impinging on an isotopically enriched self-supported ^{192}Os target. The figure highlights the difference in spectral resolution response between the HPGe and $\text{LaBr}_3(\text{Ce})$ detectors of ROSPHERE with the superior resolution of the HPGe components clearly evident. The energy resolution of the $\text{LaBr}_3(\text{Ce})$ detectors is however, in many cases sufficient to isolate specific decay sequences using the gamma-ray energy coincidence method as discussed in Section 2. This is demonstrated in Figure 6 which show background subtracted energy coincidence gates on the decays from the first $I^\pi=2^+$ and $I^\pi=4^+$ states in in ^{192}Os .

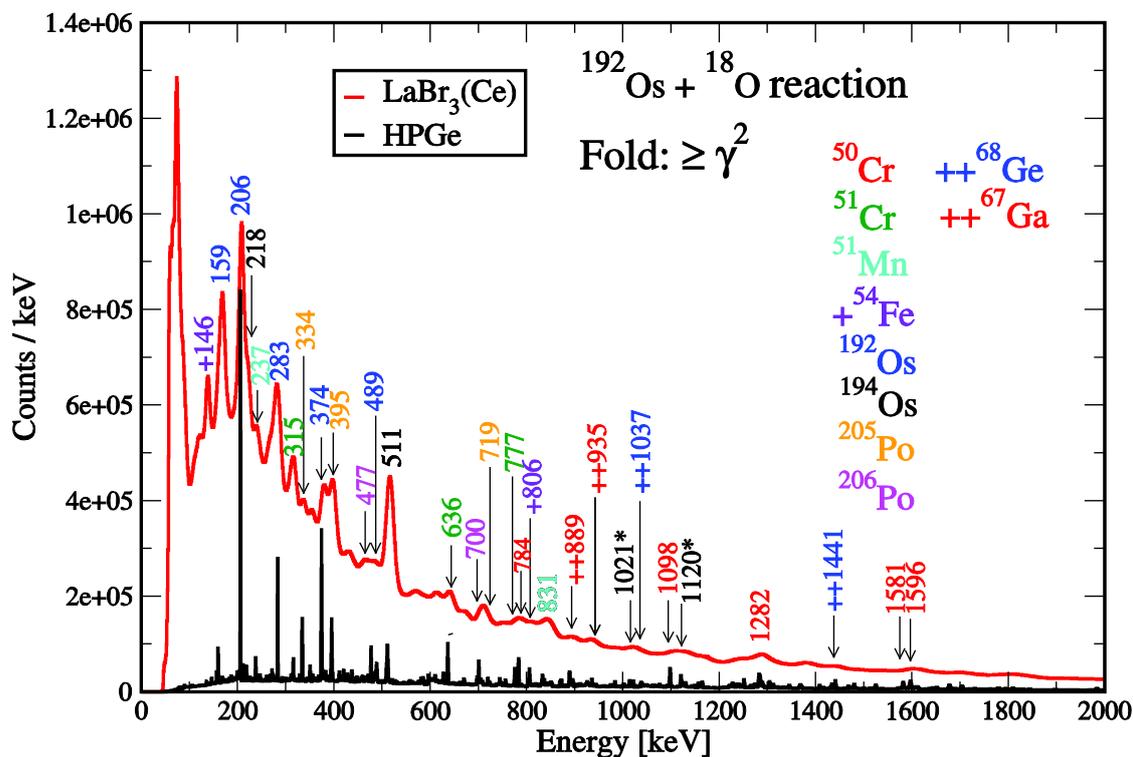


Figure 5 Total projections of the HPGe and LaBr₃(Ce) gamma-ray coincidence spectra from the $^{18}\text{O}+^{192}\text{Os}$ reaction performed at IFIN-HH using the ROSPHERE spectrometer. Discrete peaks are labelled in the HPGe spectra following reactions on the main target material and also following fusion of the ^{18}O beam with target contaminants associated with ^{28}Si , ^{40}Ca and ^{54}Fe .

Figure 6 shows a time difference spectra obtained by gating on the LaBr₃(Ce) detector full energy peaks at 206 and 374 keV in ROSPHERE. These correspond to the discrete transitions which feed into and out of the $I^\pi=2^+$ yrast state in ^{192}Os [30]. The half-life of this excited nuclear state can then be extracted from these data by evaluating the centroid of the displaced time-difference distributions forward and backward gated in time. The lifetime can also be obtained by fitting the time-difference distribution to a function which is a convolution of the Gaussian prompt response function (PRF) for the LaBr₃(Ce) detectors (which is energy dependent) together with a single exponential decay lifetime associated with the decay of the state of interest. Figure 6 shows both of these analysis methods for two sets of gates which feed into and out of the yrast $I^\pi=2^+$ state in ^{192}Os and show results for intrinsic state lifetime which are all consistent with the evaluated value for this state of 288(4) ps [30].

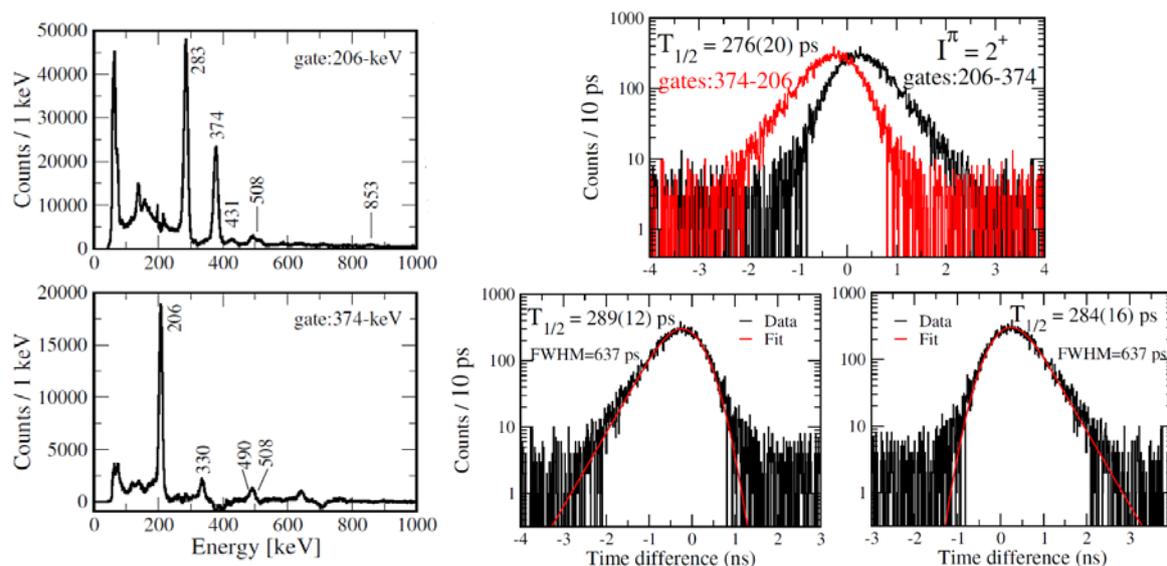


Figure 6 : (Left) Background subtracted gamma-ray coincidence spectra from gated on the $2^+_1 \rightarrow 0^+_1$ (206 keV) and $4^+_1 \rightarrow 2^+_1$ (374 keV) transitions in ^{192}Os [28] populated in unsafe Coulomb excitation following the $^{18}\text{O}+^{192}\text{Os}$ reaction. (Right) Background subtracted time-difference spectra gated between the 206 and 374 keV transition which feed into and out of the yrast $I^\pi=2^+$ state in ^{192}Os respectively. The extracted values for the lifetime of this state are consistent with the evaluated value of 288(4) ps [30]. The fit assumes a single exponential decay half-life convoluted with a Gaussian prompt response function with a full-width half-maximum of 637 ps.

4. Summary and Conclusions

The application of $\text{LaBr}_3(\text{Ce})$ detectors for nuclear spectrometry has been reviewed with emphasis on the potential uses of gamma-ray coincidence measurements for both radioactive waste assay and nuclear structure research. The NANA spectrometer which is housed at the UK's National Physical Laboratory utilised instrumentation and data analysis methodologies developed in the nuclear structure research field and will provide a dedicated national facility for radionuclide assay, radionuclide standardisations and nuclear data measurements in the coming decade.

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