



## Abstract

Beta-gamma coincidence methods have become an integral part of the International Monitoring System (IMS), particularly with regards to radionuclide detection. The UK Radionuclide Laboratory (GBL15) is currently working with partners at the University of Surrey and National Physical Laboratory (NPL) in developing a laboratory-based system for beta-gamma coincidence measurements, aiming to reduce detection limits for radionuclides of interest, as well as future-proof the UK Noble Gas analysis capability in support of the Verification Regime.

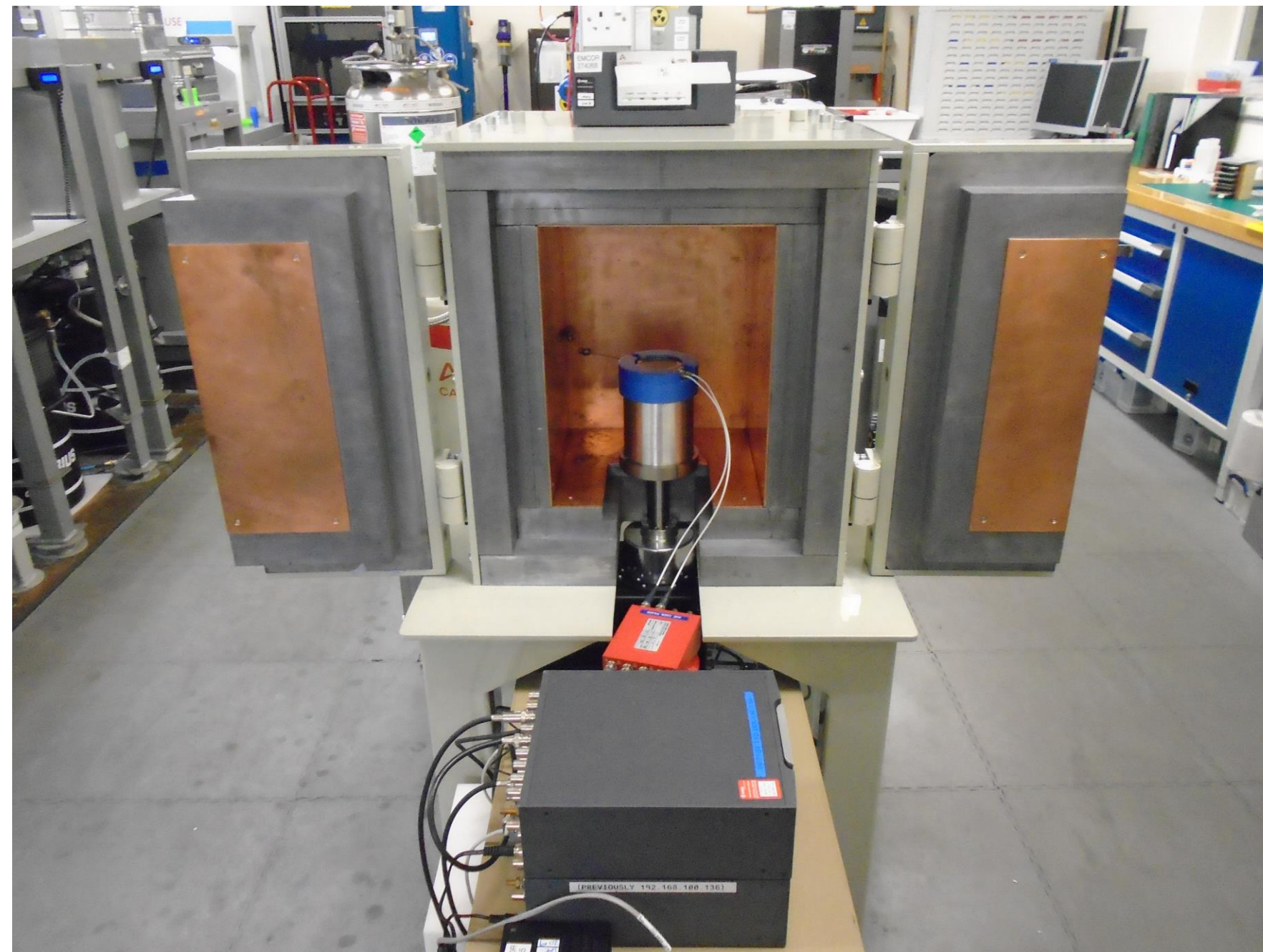


Figure 1. Prototype beta-gamma coincidence setup at GBL15, including a HPGe detector, PIPSBox and Mirion LYNX MCA

## Motivation

High resolution spectroscopy allows for better peak discrimination, less interference, better sensitivity and lower uncertainties. GBL15 are involved in the development of a number of high fidelity HPGe coincidence systems, reaching ultra-low detection limits [1]. A future noble gas detector system could be suited to the laboratory, which contains a suite of HPGe detectors, offering room for expansion if required. Developing the system in-house is helping to sustain growth of skills in noble gas radiometry at AWE, where a new facility is planned for GBL15 within the next 5 years. As such, a full options study is required to assess the viability of future noble gas laboratory and the technologies it may use.

Whilst developing a new system in-house offers many benefits, such as a full understanding of the system without any 'black box' technology, noble gas measurement systems require gas handling equipment in order to prepare the radionuclide for radiation measurement. Such equipment has not been developed at GBL15 previously, so must be considered as part of this work.

Mirion provide an aluminium & carbon fibre gas cell containing two PIPS (Passivated Implanted Planar Silicon) detectors, which are capable of measuring the  $\beta$  particles emitted as a result of the decay of radionuclide fission product isotopes  $^{133}\text{Xe}$  and  $^{135}\text{Xe}$ . The PIPS detectors can also measure the (discrete energy) internal conversion electrons emitted from the meta-stable isotopes  $^{131\text{m}}\text{Xe}$  and  $^{133\text{m}}\text{Xe}$  as they transition to lower energy levels. Being able to resolve the energy of these electrons is an important part of  $\beta$ - $\gamma$  (electron-photon) radionuclide measurements.

Performing these measurements in coincidence with high-purity germanium constitutes a high energy resolution  $\beta$ - $\gamma$  system and forms the basis of this study.

## Measurements at AWE

Radionuclide samples provided by Seibersdorf Laboratories were measured on the GBL15 working prototype system in January 2019. The efficiency of the transfer of xenon from container to PIPSBox was measured via gamma spectrometry before and after transfer and the dead volume of the system had been previously evaluated by pressure differential.

The aim of this measurement campaign was a proof of principle for the laboratory to experience high resolution beta-gamma coincidence spectrometry and the learn from this system to assess the prospect of using PIPSBox technology at GBL15. Some of the results are given below.

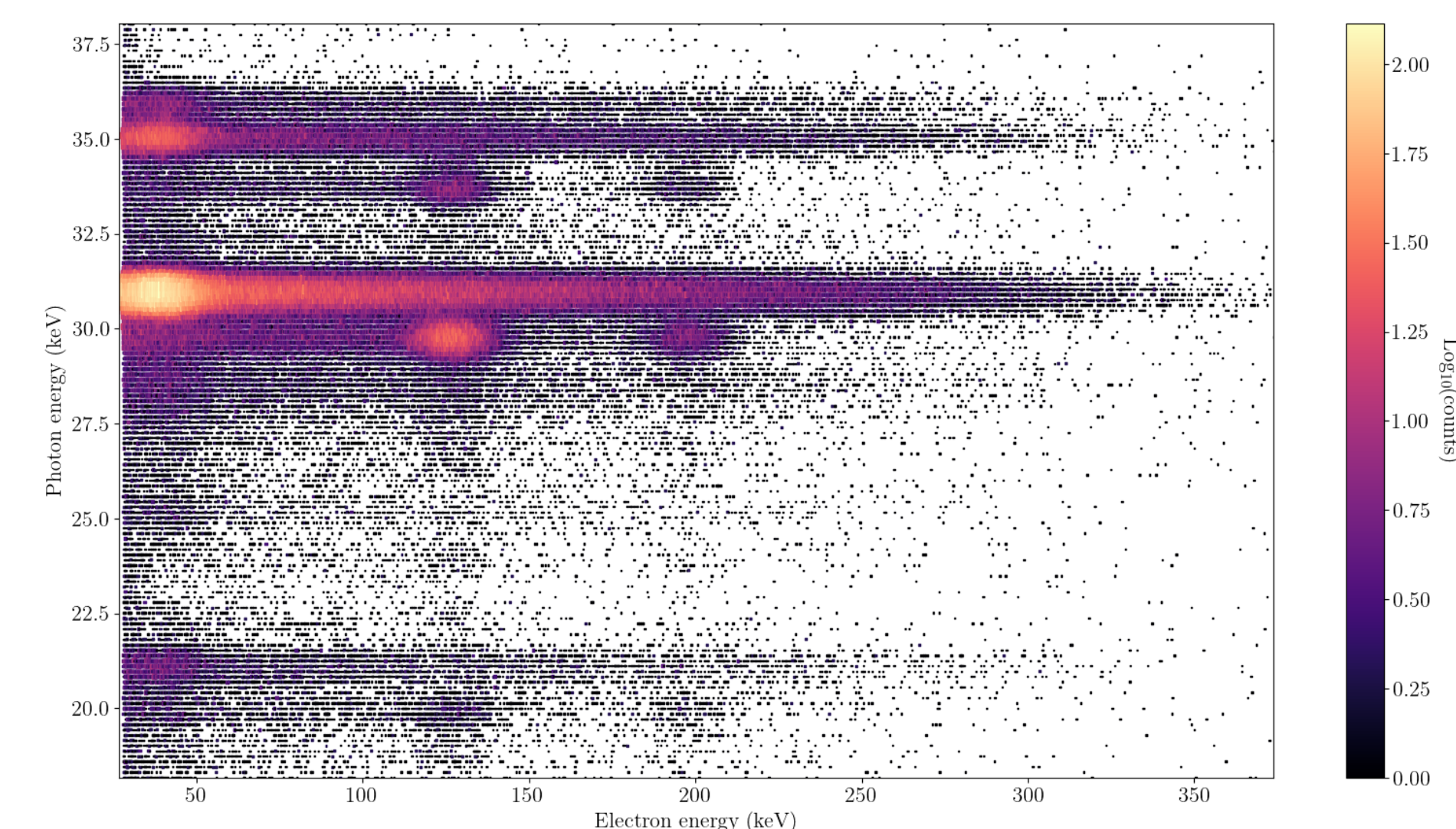


Figure 2. Electron-photon coincidence spectrum for the measurement of a multi isotope radionuclide sample. Conversion electrons and beta continua are present and clearly resolved

Table 1. Results of measurements made on the prototype system, compared to reference values. Calibrations performed by measurement

Isotope	Region	Counts	CPS	K	Abundance	$\beta$ - $\gamma$ Efficiency (%)	Activity (Bq)	Ratio to Reference
$^{131\text{m}}\text{Xe}$	5	8443	0.04956	1.45	0.442	3.50	12.04	1.136
$^{131\text{m}}\text{Xe}$	7	2395	0.0141	1.45	0.103	4.37	11.76	1.109
$^{133\text{m}}\text{Xe}$	6	952	0.00559	6.49	0.453	3.52	5.902	1.357
$^{133\text{m}}\text{Xe}$	3	56150	0.330	2.21	0.37	5.91	86.68	1.043

By using a first measurement to generate efficiencies for coincidence regions of interest (ROI), the second measurement was able to be quantified. Table 1 shows the results when compared to the reference value. More work is required to fully account for uncertainties, assess detection limits and develop a rigorous calibration procedure.

## Prototype Measurement System at NPL

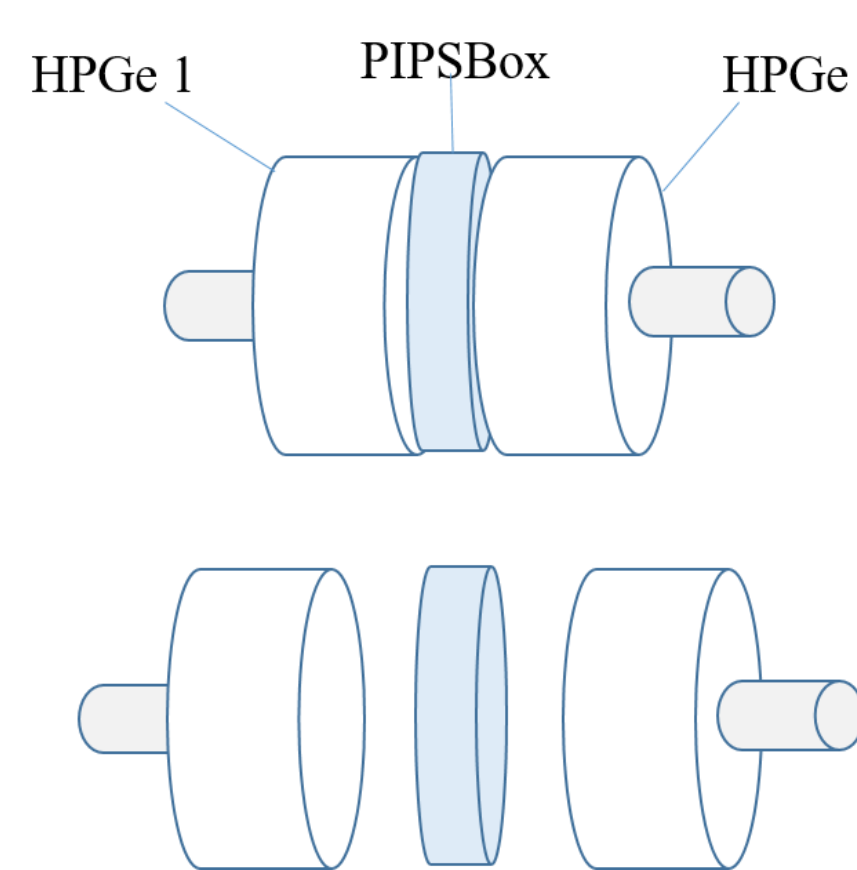


Figure 3. AWE Detector setup at NPL

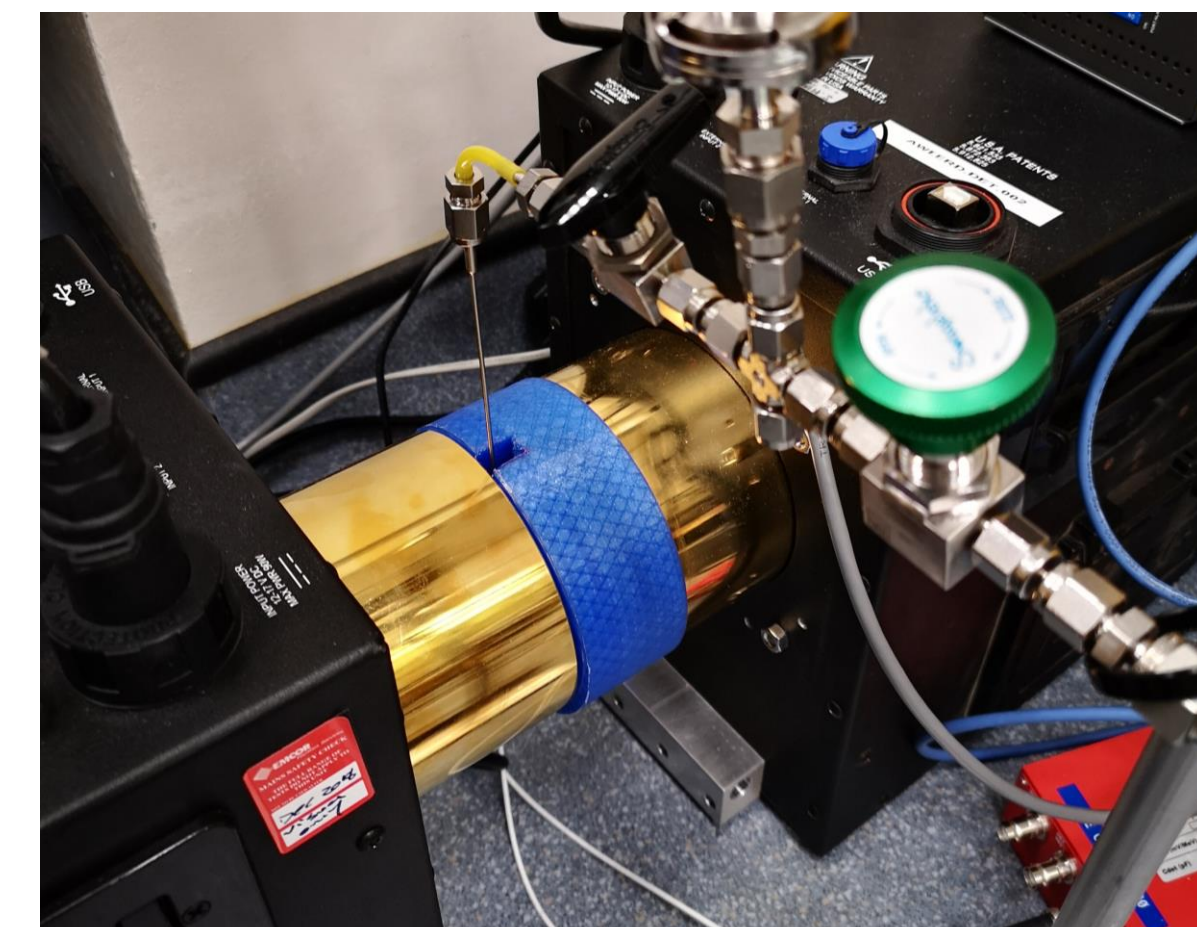


Figure 4. PIPSBox-HPGe setup at NPL, for measurement of prompt fission gases

## Measurements at NPL

NPL (National Physical Laboratory, UK) have developed a gas processing system for the extraction of fission gases from a custom developed HEU target. The target has been irradiated at the NPL neutron irradiation facility and the gas extracted for measurement within one hour. A portion of this prompt fission gas was transferred into a portable HPGe-PIPSBox detector system (see figures 3-4).

The first extraction provided a gas sample containing  $^{85\text{m}}\text{Kr}$ ,  $^{87}\text{Kr}$ ,  $^{88}\text{Kr}$ ,  $^{135}\text{Xe}$  &  $^{133}\text{Xe}$ . A second extraction of the same irradiated target (one week later) provided a mixed radionuclide sample containing  $^{133}\text{Xe}$ ,  $^{133\text{m}}\text{Xe}$ ,  $^{135}\text{Xe}$  and  $^{133}\text{Xe}$ ; an ideal gas mixture for CTBT verification measurements. Utilising two HPGe detectors in a close geometry made it possible to measure  $\gamma$ - $\gamma$  coincident signatures for a number of gaseous radionuclides, such as that shown in figure 7.

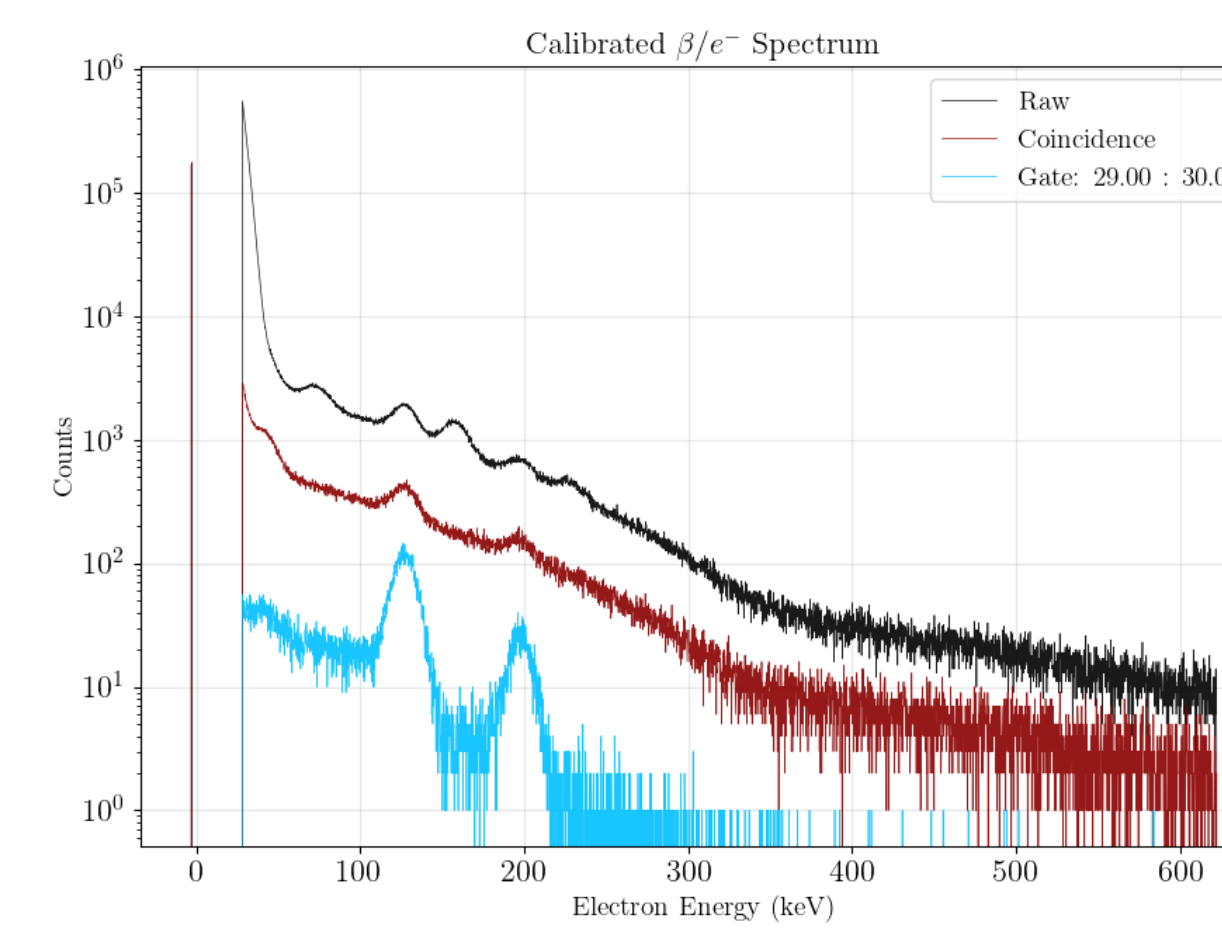


Figure 5. Electron/Beta spectrum from the measurement of a mixed isotope radionuclide sample. Total coincidence (red) and selected coincidence (X-ray energy, blue) show the  $^{131\text{m}}\text{Xe}$  and  $^{133\text{m}}\text{Xe}$  129 keV and 199 keV electrons clearly.

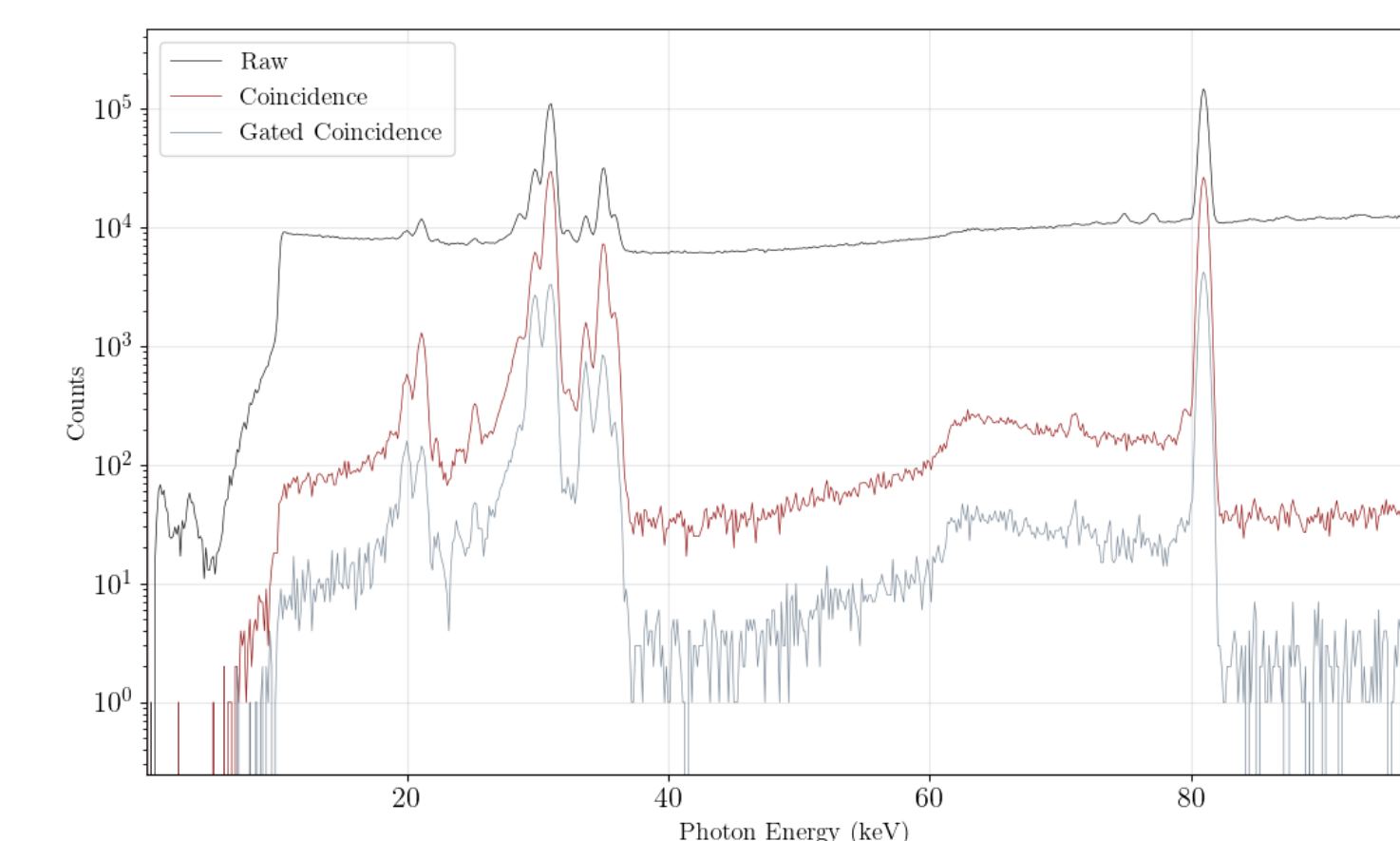


Figure 6. X/gamma spectrum from the measurement of a mixed radionuclide sample. X-rays from Xe-133 and Xe-135 are present at 29-36 keV. The corresponding Germanium escape peaks are present at ~20 keV. Xe-133 gamma can be seen at 81 keV. Using the  $\beta/e$  signal to gate the gamma greatly improves the resolution of the X-rays, helping to de-convolute them from one another.

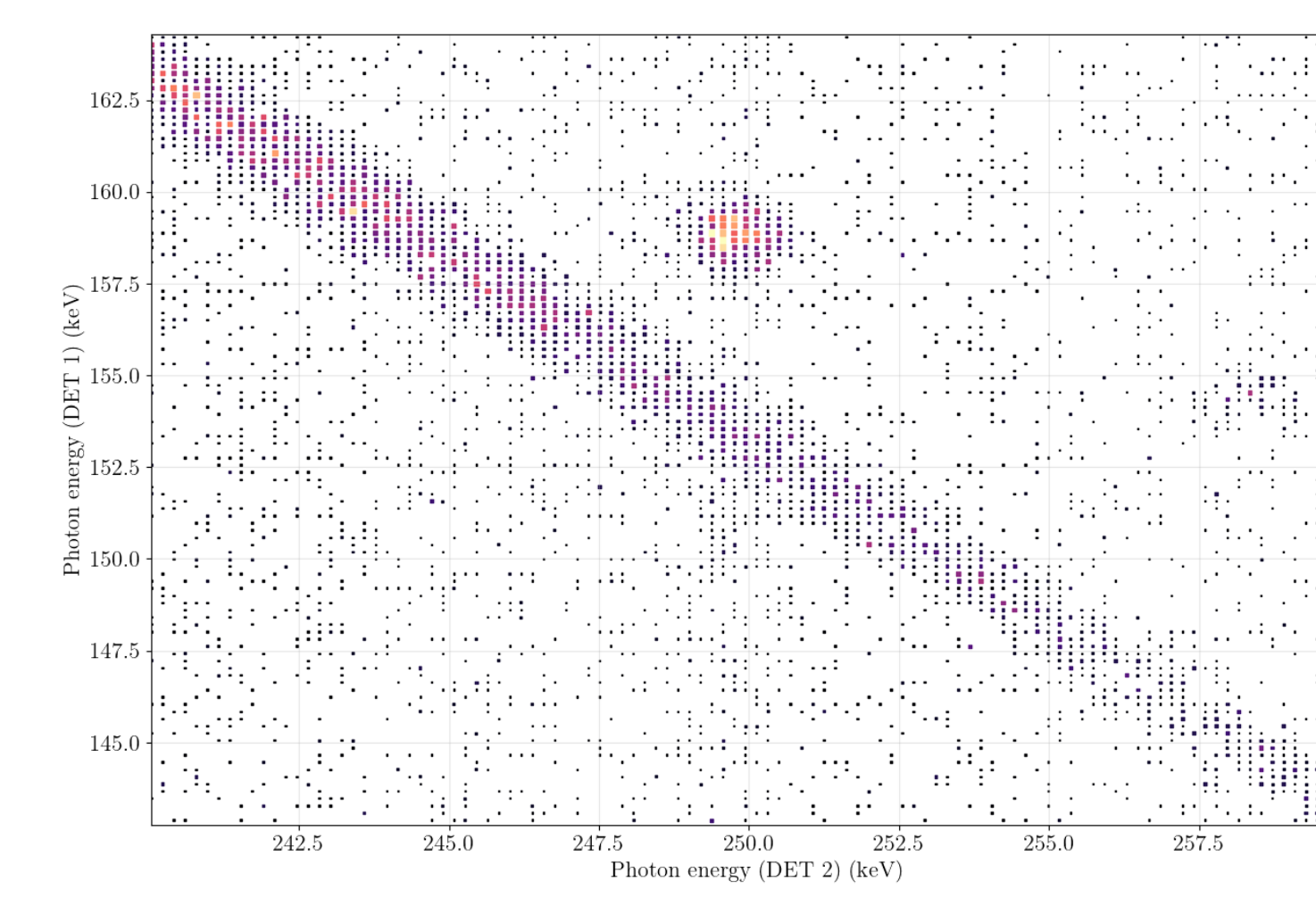


Figure 7.  $\gamma$ - $\gamma$  coincidence spectrum showing the Xe-135 coincidence between 158 keV and 250 keV. The spectrum is beta-gated to reduce the background counts, thus giving a  $\beta$ -gated  $\gamma$ - $\gamma$  matrix.

## Findings and Results

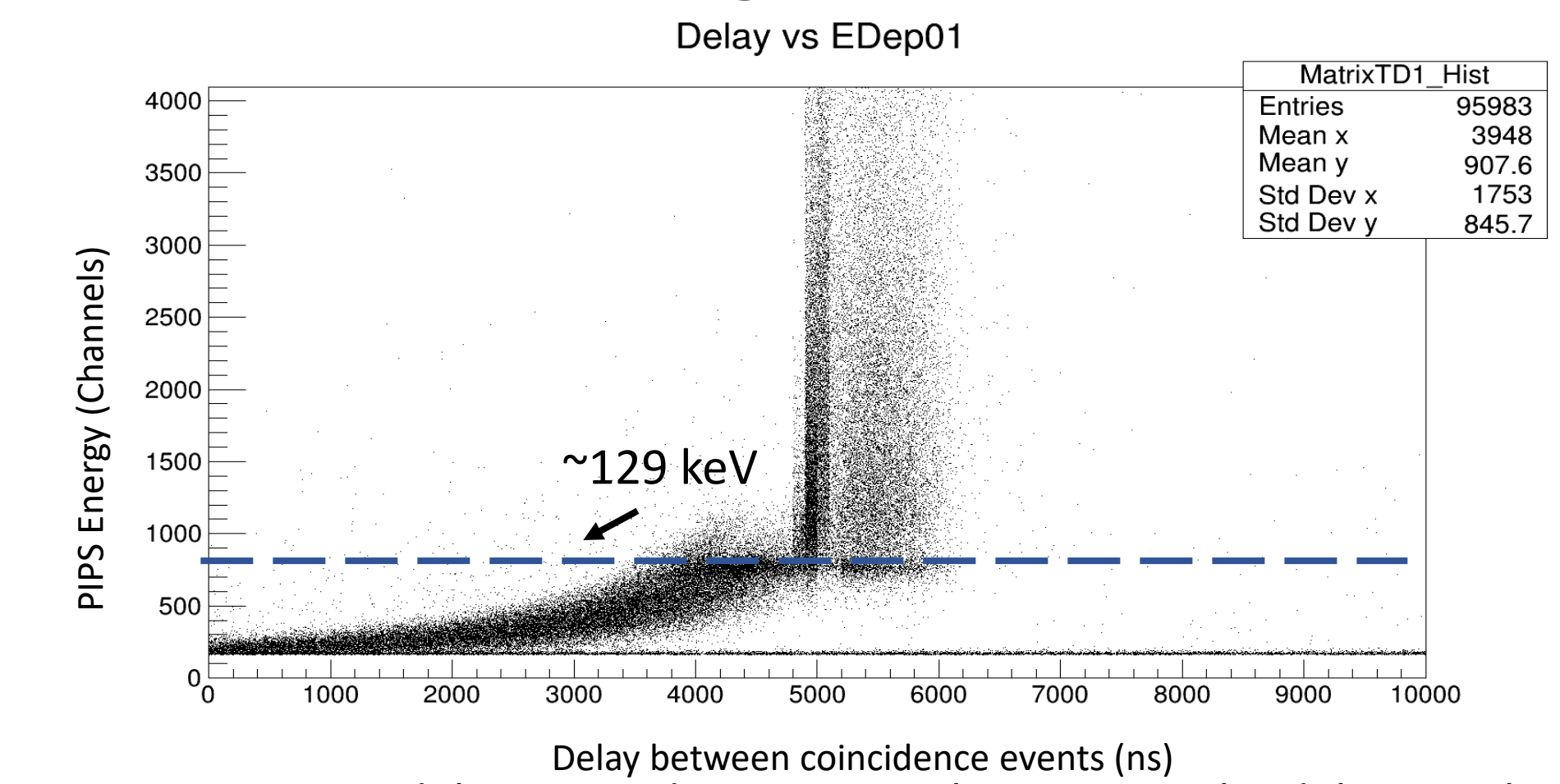


Figure 8. Timing delay-coincidence energy histogram. The delay is calculated as  $t[\text{PIPS}] - t[\text{HPGe}]$  where  $t$  is the time of the event in each detector.

Performing these measurements on a custom system produced some unexpected outcomes. A particular highlight includes the Delay-Coincidence energy spectrum collected from the measurement of  $^{131\text{m}}\text{Xe}$  in a low background environment. This system comprised of one BEGe detector and a single PIPSBox detector. The coincidences between the two detectors were used to populate the usual 2D matrices, and the timing between the events was also considered. From Figure 8 it is clear that as the energy deposited in the silicon decreases from 129 to ~0 keV, the delay between the two detections decreases from ~5  $\mu\text{s}$  to 0.

Projecting the electron-X-ray spectrum for 4 arbitrary regions of the delay spectrum illustrates the process of electron straggling in the detector cell is responsible for the result. Where the electron interacts with the PIPS without delay, the full energy is deposited in the usual 129 keV ( $e$ ) and 29 keV ( $X$ ) region, with ~5  $\mu\text{s}$  delay as the low energy charge is collected in the HPGe. In the instances where the conversion electron interacts with air molecules and other gases in the sample, it deposits less energy and the delay is reduced as a consequence. Further work is required to understand the effect of electron straggling on the results of this technique.

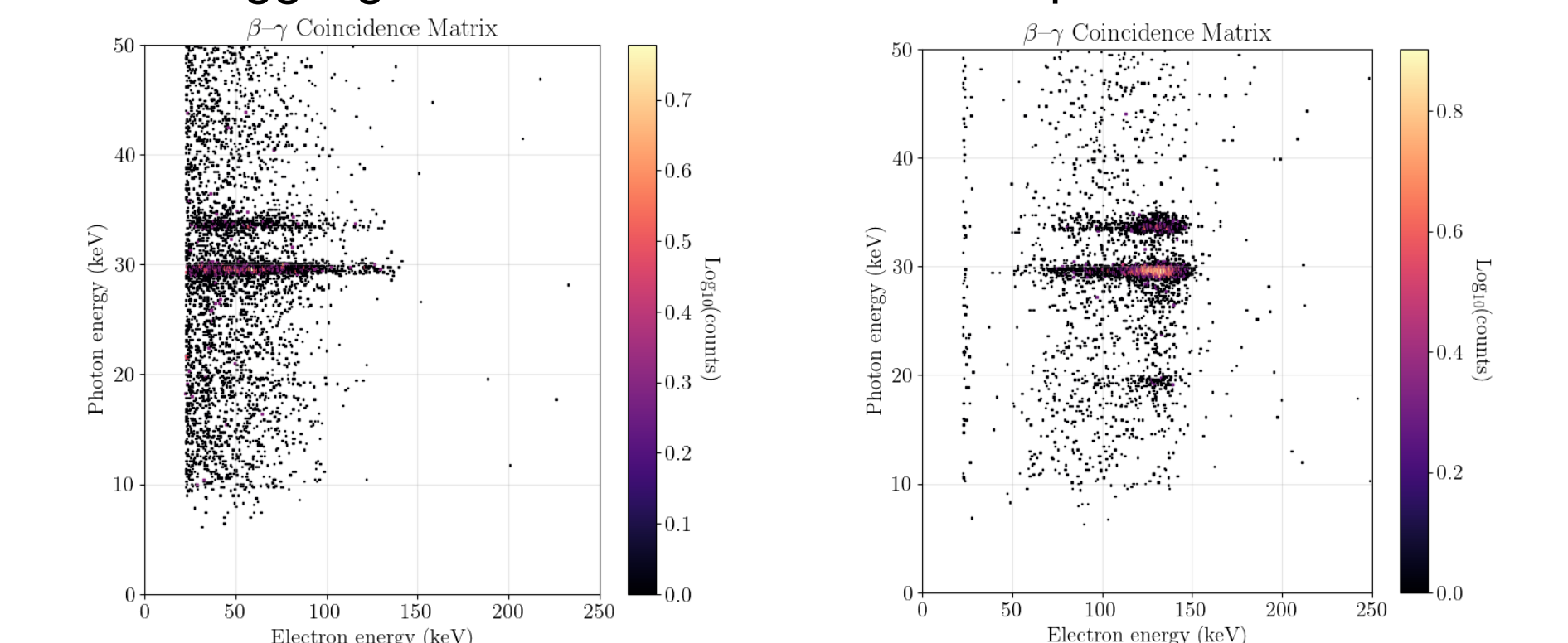


Figure 9a. Coincidence matrices for the 0-3800 ns window (left) and for the 3800-4200 ns window (right)

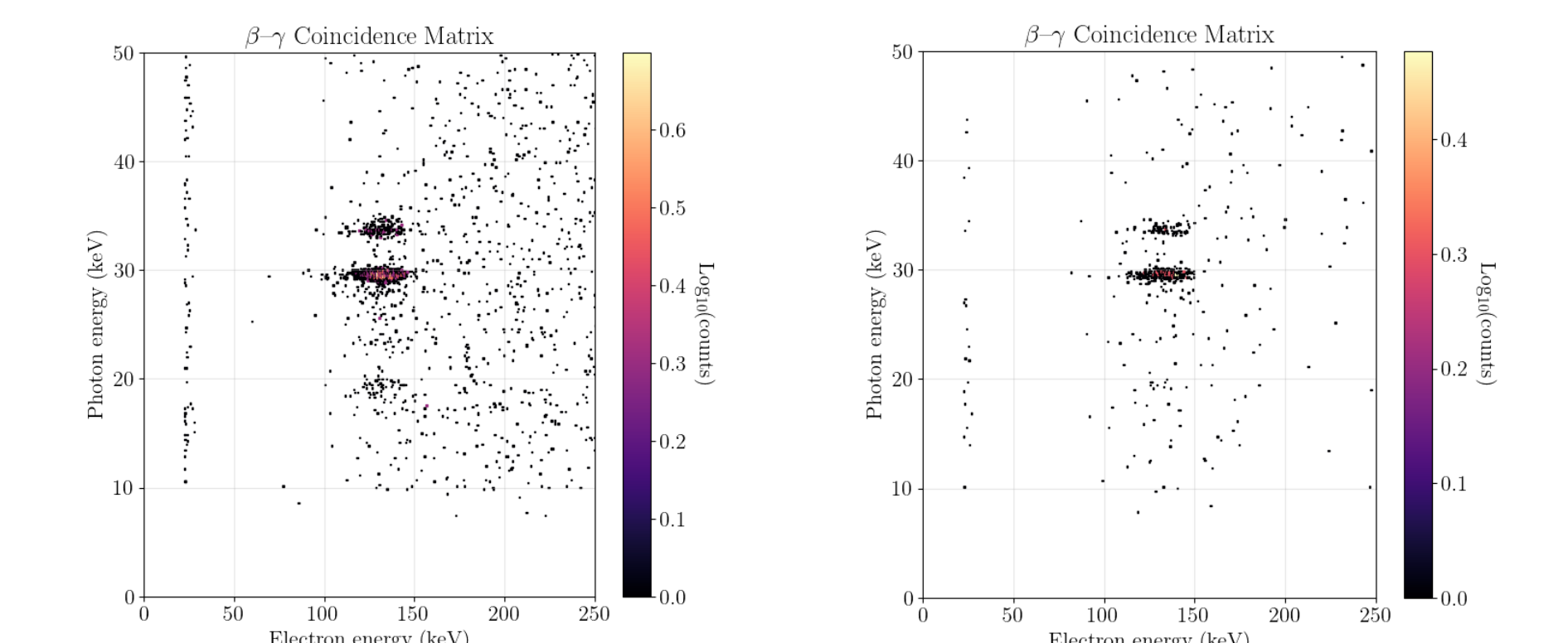


Figure 9b. Coincidence matrices for the 4200-5200 ns window (left) and for the 5200-6200 ns window (right)

The next stages of this work will include measurements using a LEGe detector and faster electronics; GEANT4 simulations and other detection media shall also be considered. By 2019 we aim to complete an options study on future noble gas systems at GBL15 and have a prototype high resolution beta-gamma laboratory system for radionuclide measurements, by the end of 2020.

[1] Britton R, Davies A, Jackson M; Quantifying radionuclide signatures from a gamma-gamma coincidence system, JER 149, 158-163