Towards Complete Decay Spectroscopy of ^{152}Tb: a Diagnostic Component of the Terbium Theragnostic Toolbox

E.B.O'Sullivan[1,2], S.M.Collins[1,2], J.M.Daugas[3], L.Domenichetti[3], J.Heery[1], J.Henderson[1], U.Köster[3], C.Michelagnoli[3], T.Parry[1], S.Pascu[1], P.H.Regan[1,2], R.Shearman[2]

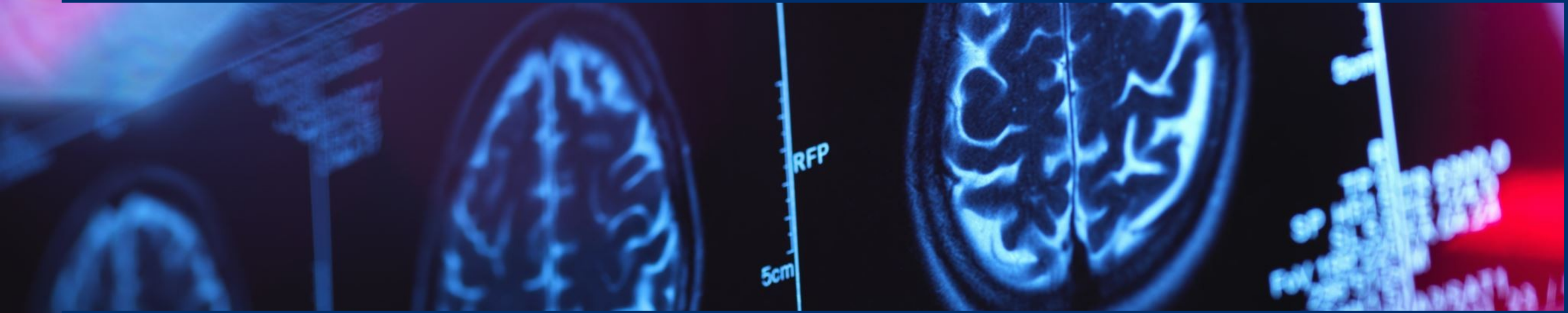
1. University of Surrey, Guildford, UK
2. National Physical Laboratory, Teddington, UK
3. Institut Laue-Langevin, Grenoble, France

IoP Conference 2025

^{152}Tb: Medical Imaging

NPL

- β^+ / EC decay to ^{152}Gd – **Positron Emitter**
- $T_{1/2}$ = 17.8784(95) h
- $I_{\beta+}$ = 20.3(15)%
- Q_{EC} = 3990(40) keV [1]
- First-in-human trials show promise in **PET imaging**: ^{152}Tb-DOTATOC and ^{152}Tb-PSMA-617 used successfully in **human patients** [2,3]
- Terbium isotope – applications in **theragnostics**



1) Nuclear Data Sheets for A = 152, M.J. Martin
2) Preclinical investigations and first-in-human application of ^{152}Tb-PSMA-617 for PET/CT imaging of prostate cancer, C. Müller et. al
3) Clinical evaluation of the radiolanthanide terbium-152: first-in-human PET/CT with ^{152}Tb-DOTATOC, R.P. Baum et. al

Theragnostics: Therapy + Diagnostics

NPL

- **Terbium theragnostic quartet**: four different medical uses
- Shared chemistry – compatible with the **same delivery mechanism**
- **Personalised medicine** – treatment plan tailored to individual patients

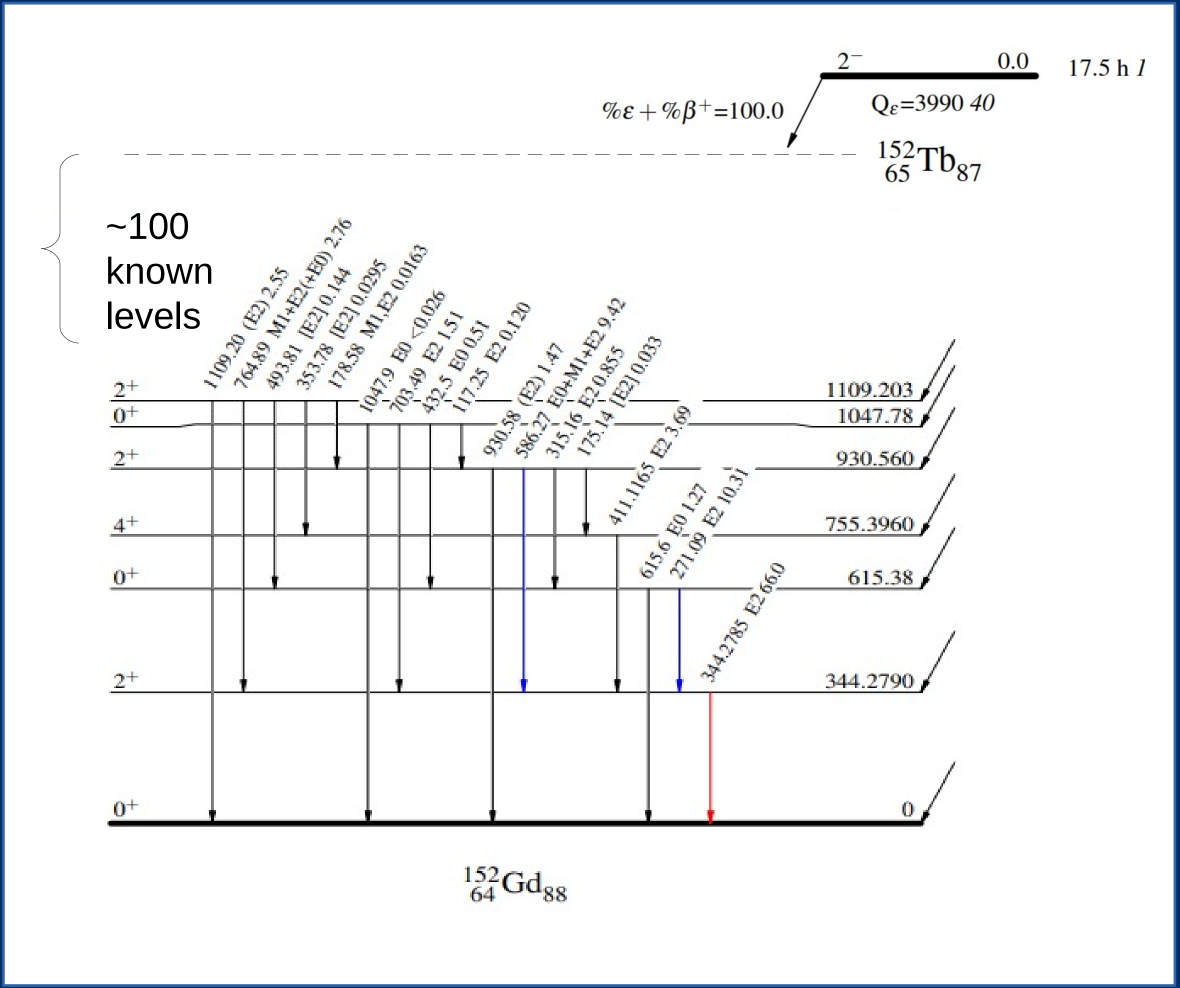
Isotope	$T_{1/2}$	Decay	Use
^{149}Tb	4.118(25) h [4]	Alpha	Radionuclide Therapy
^{152}Tb	17.8784(95) h [5]	Beta+ / EC	PET Imaging
^{155}Tb	5.2346(36) d [6]	EC	SPECT Imaging
^{161}Tb	6.9637(29) d [7]	Beta-	Radionuclide Therapy

4) PRISMAP Radionuclide Portfolio, https://www.prismap.eu/radionuclides/portfolio/149Tb
5) Determination of the Terbium-152 Half-Life from Mass-Separated Samples from CERN-ISOLDE and Assessment of the Radionuclide Purity, S.M. Collins et. al
6) Half-life determination of ^{155}Tb from mass-separated samples produced at CERN-MEDICIS, S.M. Collins et. al
7) Determination of the ^{161}Tb half-life, S.M. Collins et. al

Nuclear Data: ^{152}Gd

NPL

- ^{152}Tb → ^{152}Gd decay **last studied in 2003,** using a **pair of HPGe detectors** [8]
- Highest energy level identified at 3358 keV – **600 keV below Q_{EC}**
- 248 out of 635 known transitions **unplaced**
- RIPL-3 level density calculation – **400 states** with $J^{\pi}=1^{-},2^{-},3^{-}$ predicted in range 3-4 MeV
- Pandemonium effect: unknown high energy states leads to **inaccurate beta dose**, for example in ^{86}Y [9].

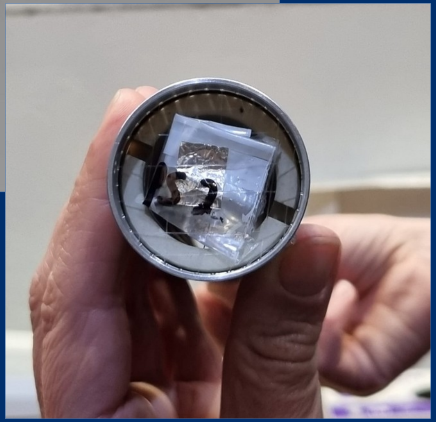


8) Properties of ^{152}Gd Collective States, J. Adam et al.
9) RIPL — Reference input parameter library for calculation of nuclear reactions and nuclear data evaluations, R. Capote et al.
10) State-of-the-art γ-ray assay of ^{86}Y for medical imaging, A.C. Gula et. al.

^{152}Tb Decay Spectroscopy

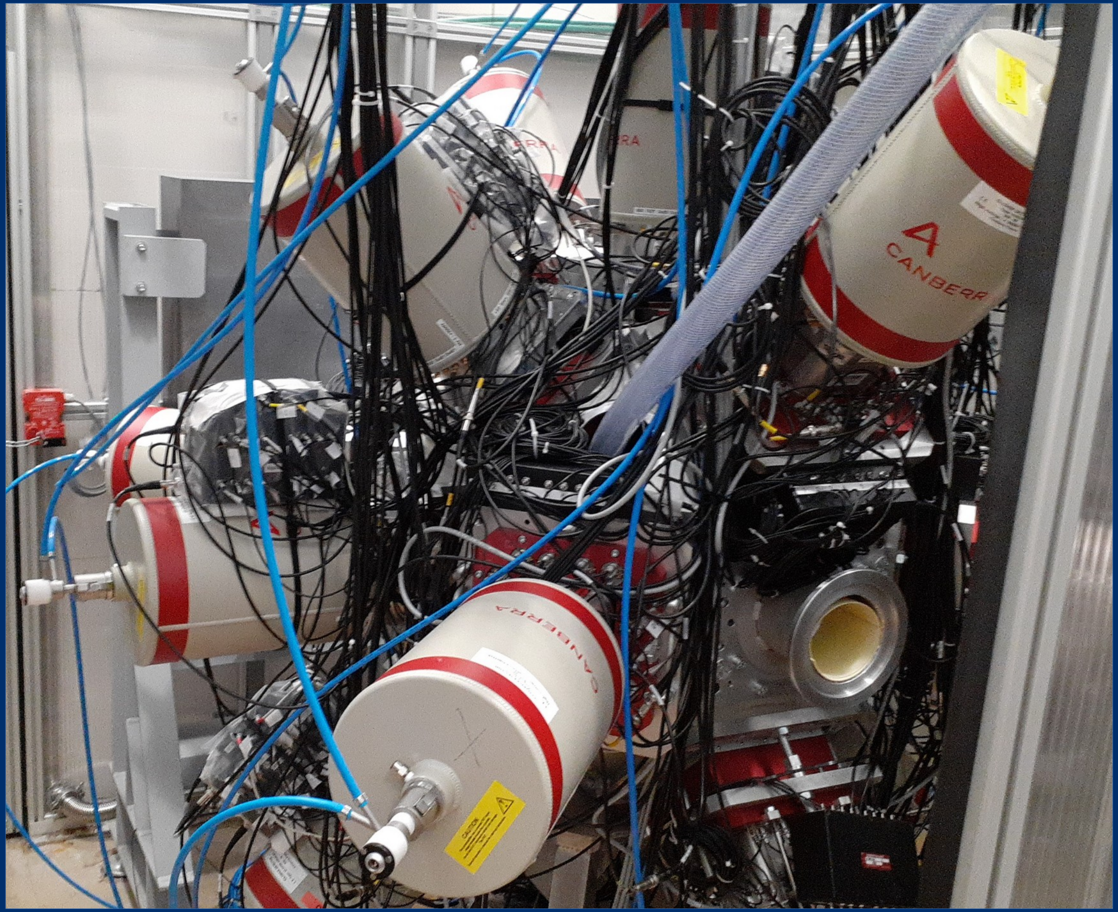
NPL

- Sources prepared at CERN ISOLDE: **1.4 GeV proton beam** on a tantalum target
- Samples purified by **laser ionisation** and **mass separation** and implanted onto a pair of Al foils plus one Mylar foil
- Delivered to ILL Grenoble for measurement: **1×10^5 Bq and 5×10^5 Bq** at start of experiment (3rd May 2023)



Gamma-Gamma Spectroscopy

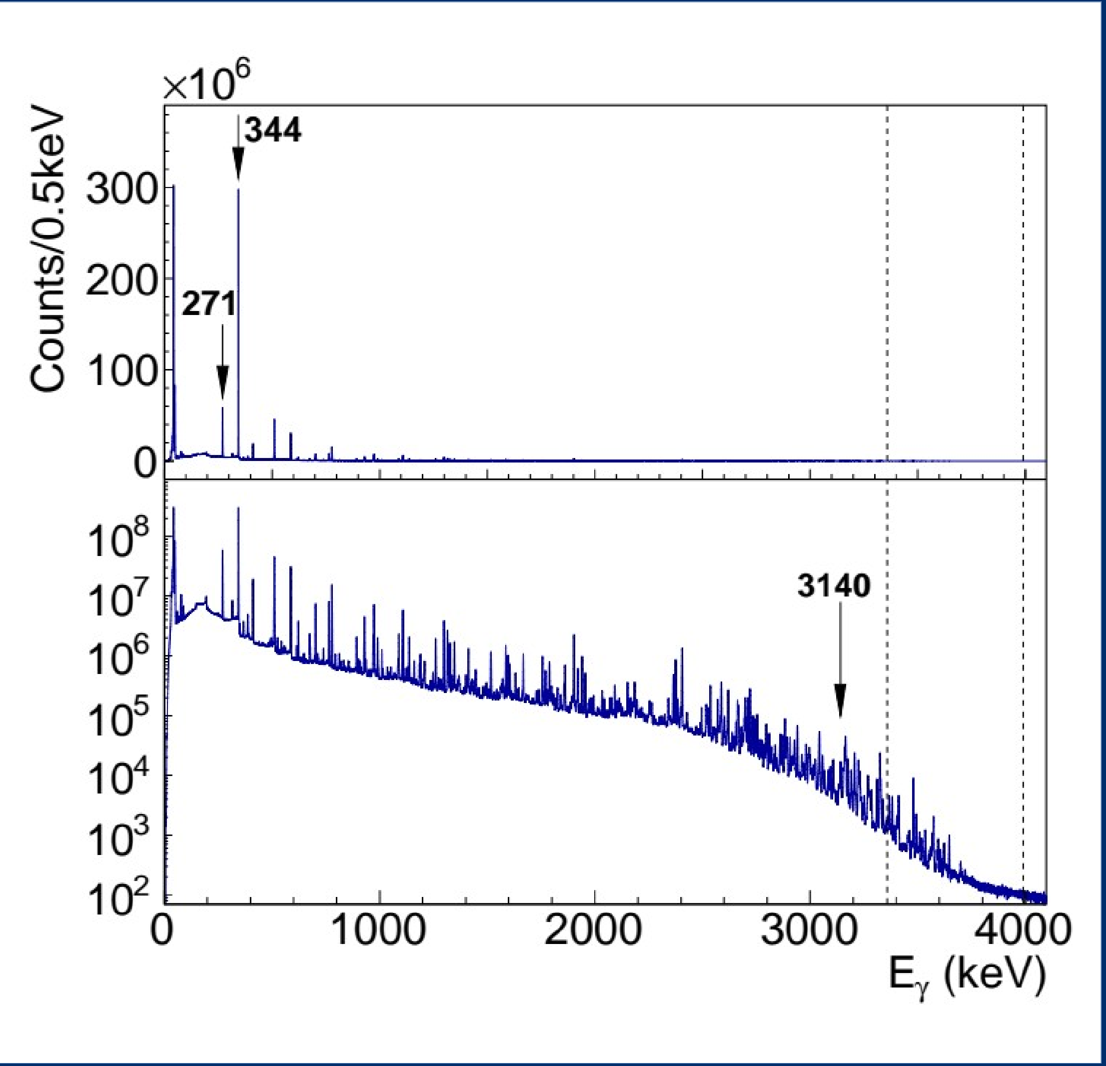
- FIssion Product Prompt Gamma-Ray Spectrometer (FIPPS) [10]
- **64 HPGe crystals**, 16 clovers with BGO shielding (14 crystals excluded)
- Absolute efficiency ~9% at 344 keV
- **7 days** measurement time
- Electron-Gamma spectroscopy carried out in parallel – PN1/LOHENGRIN



11) FIPPS (FIssion Product Prompt γ-ray Spectrometer) and its first experimental campaign, C. Michelagnoli et. al

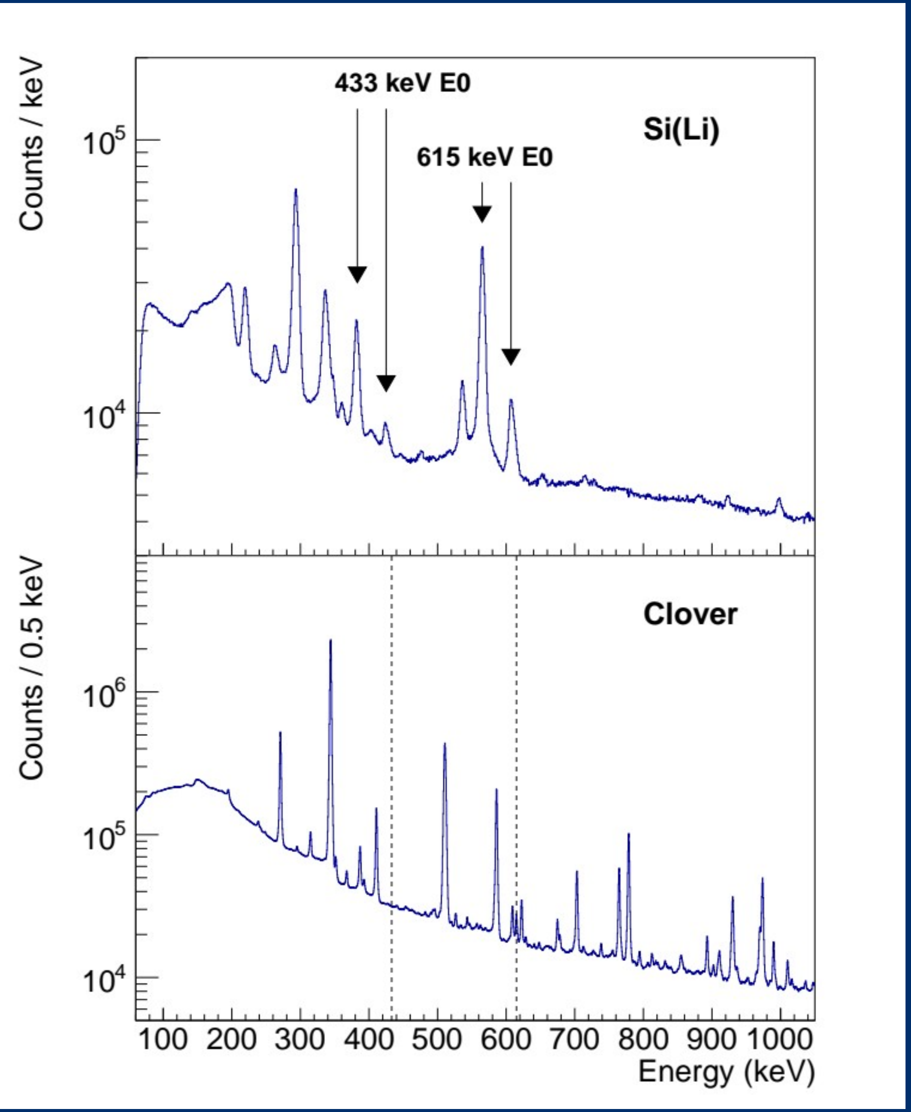
Singles Spectra

- **1.5e10 single events** collected from source 2 (~70% of the total)
- Highest energy gamma previously placed: 3140 **keV**
- Highest energy state previously identified: **3358 keV**
- Q_{EC} = **3990 keV**



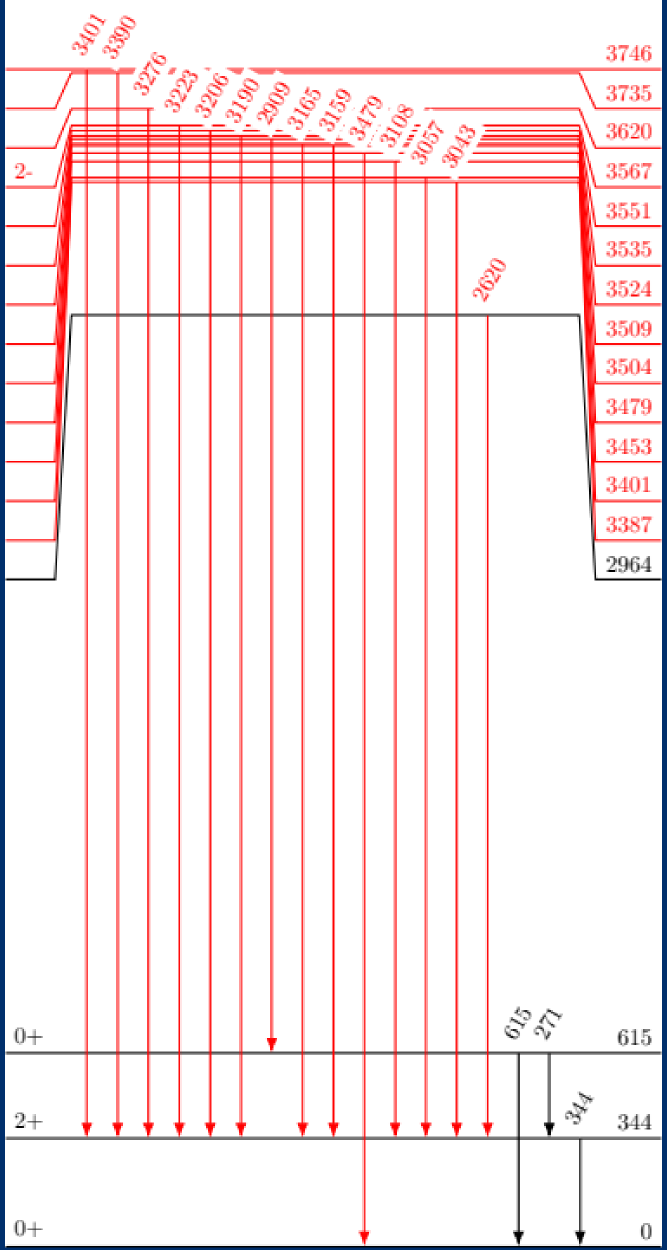
Electron-Gamma Spectroscopy

- Internal conversion – **electrons emitted** instead of gammas
- **Invisible** to HPGe array
- Electron energy **depends on orbital** – K & L peaks
- 0+ to 0+ transitions – no single-gamma decay mode, E0 transitions **only emit electrons**



Preliminary Results

- 13 previously unidentified states reported so far following gamma-gamma coincidence analysis
- Relative intensities of E0 transitions measured, and K/L electron intensity ratios used to validate BrICC predictions

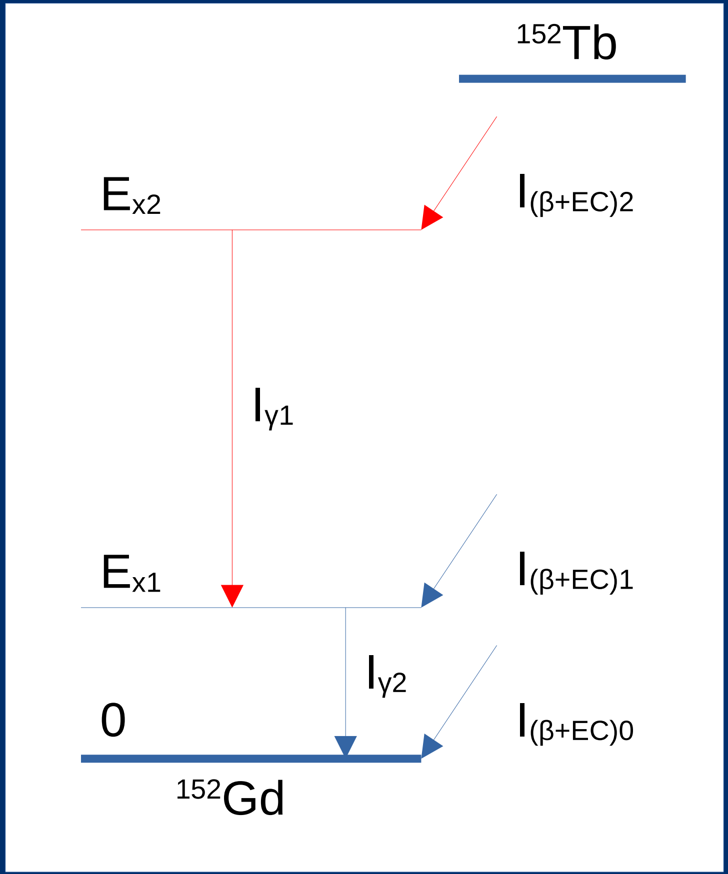


12) Towards complete decay spectroscopy of 152Tb, E.B. O’Sullivan et al.
13) Electron-gamma spectroscopy of 152Tb, E.B. O’Sullivan et al., submitted

Level Feeding

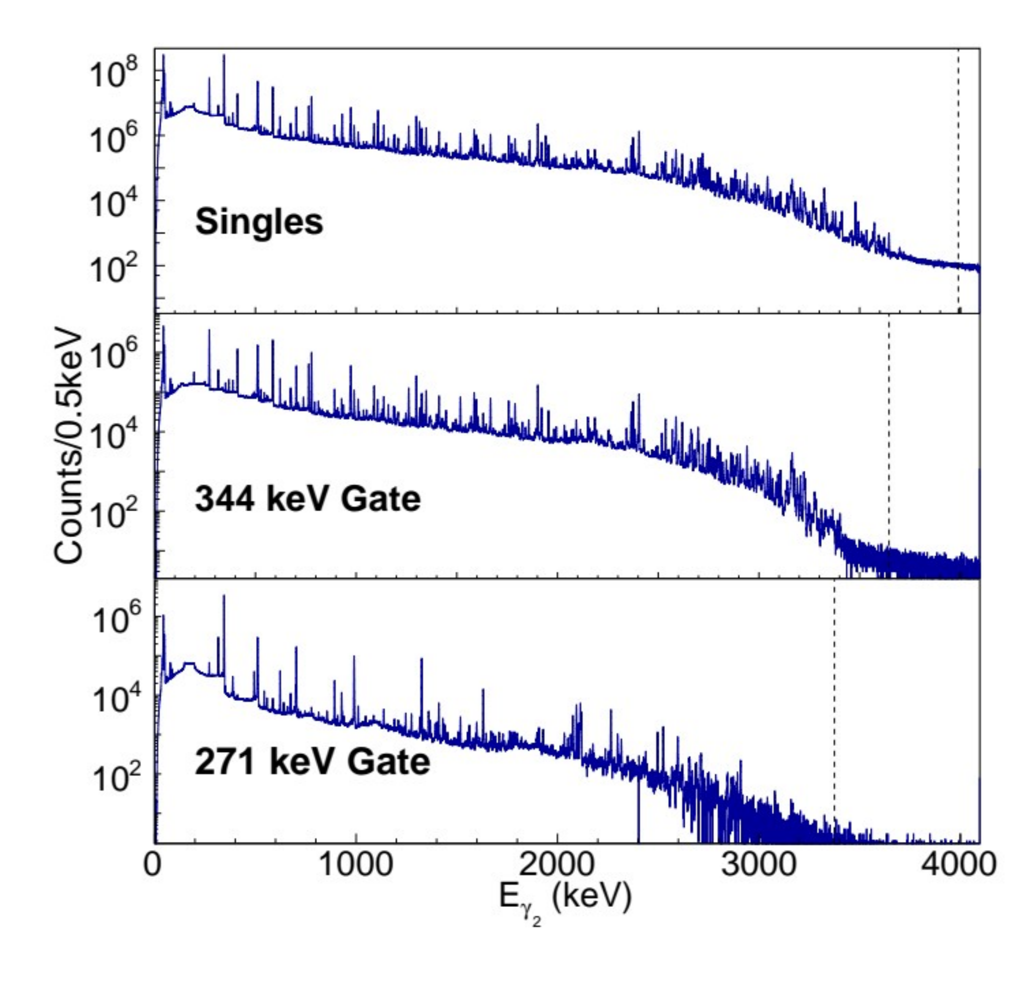
NPL

- Investigate transitions from 13 new states to ground + first 5 excited states
- Assume all gamma ray intensity replaces feeding to low-energy states – 0.33% of total so far
- 344-keV 2+ state: 1.9% of feeding replaced
- 755-keV 4+ state: 8.2% of feeding replaced
- Angular momentum selection rules: decay to both states first forbidden
- β^+/EC feeding: investigate coincidences with 511-keV and x-rays



Future Work

- Gamma-gamma coincidence analysis – continue identifying states + placing transitions
- Angular correlation analysis – spin/parity assignments
- Electron-gamma coincidence analysis – E0 strengths and ICC
- Monte Carlo simulations – validate efficiency curve for intensity measurements
- Balance final intensities to derive beta feeding
- Support with complementary techniques (e.g. TAGS) – high efficiency alone does not fully mitigate pandemonium effect



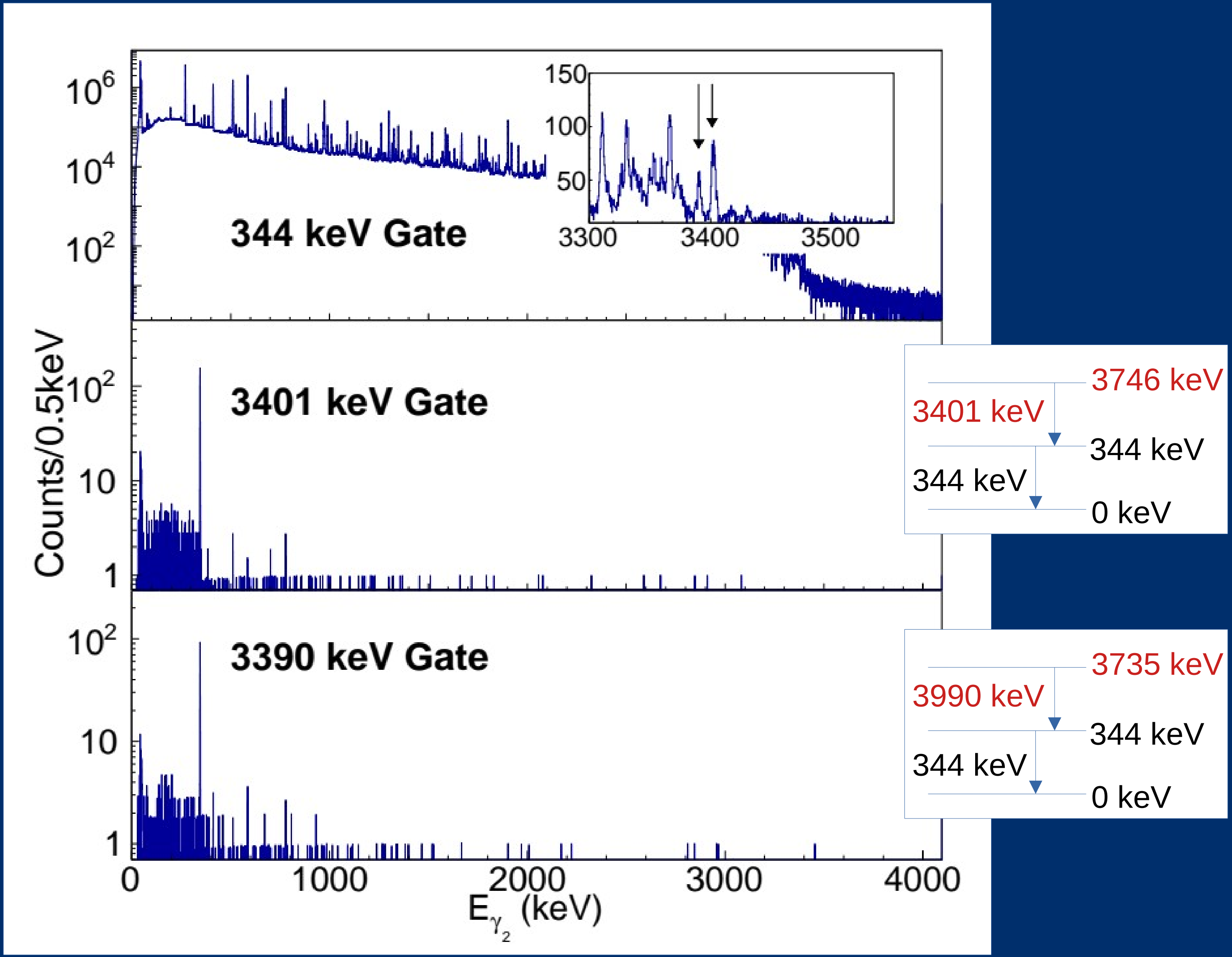
References:

1. Nuclear Data Sheets for A = 152, M.J. Martin, Nuclear Data Sheets 114 (2013) 11
2. Preclinical investigations and first-in-human application of ^{152}Tb-PSMA-617 for PET/CT imaging of prostate cancer, C. Müller et al, EJNMMI Research 9 (2019) 68
3. Clinical evaluation of the radiolanthanide terbium-152: first-in-human PET/CT with ^{152}Tb-DOTATOC, R.P. Baum et al, J. Nucl. Med. 53 (2012) 12
4. PRISMAP Radionuclide Portfolio, https://www.prismap.eu/radionuclides/portfolio/149Tb
5. Determination of the Terbium-152 Half-Life from Mass-Separated Samples from CERN-ISOLDE and Assessment of the Radionuclide Purity, S.M. Collins et al, Appl. Radiat. Isot. 202 (2023) 111044
6. Half-life determination of ^{155}Tb from mass-separated samples produced at CERN-MEDICIS, S.M. Collins et al, Appl. Radiat. Isot. 190 (2022) 110480
7. Determination of the ^{161}Tb half-life, S.M. Collins et al, Appl. Radiat. Isot. 182 (2022) 110140
8. Properties of ^{152}Gd Collective States, J. Adam et al, EPJA 18 (2003) 65
9. RIPL — Reference input parameter library for calculation of nuclear reactions and nuclear data evaluations, R. Capote et al., Nuclear Data Sheets 110 12 (2009), 3107–3214
10. State-of-the-art γ-ray assay of ^{86}Y for medical imaging, A.C. Gula et al. Phys. Rev. C 102 (2003) 034316
11. FIPPS (FIssion Product Prompt γ-ray Spectrometer) and its first experimental campaign, C. Michelagnoli et al, EPJ Web Conf. 193, (2018) 04009
12. Towards complete decay spectroscopy of 152Tb, E.B. O’Sullivan et al, Radiation Physics and Chemistry 232 (2025) 112641
13. Electron-gamma spectroscopy of 152Tb, E.B. O’Sullivan et al, Physica Scripta, submitted

EOS acknowledges funding via a PhD scholarship bursary from the EPSRC. The work performed at NPL was supported by the National Measurements System Programmes Unit of the UK’s Department for Science, Innovation and Technology. PHR also acknowledges funding from the UK Science and Technologies Facilities Council under grant numbers ST/P005314/1, ST/V001108/1, ST/L005743/1 and ST/ P005314. Thank you to the CERN-ISOLDE teams, including target, RILIS, operations, and RP.

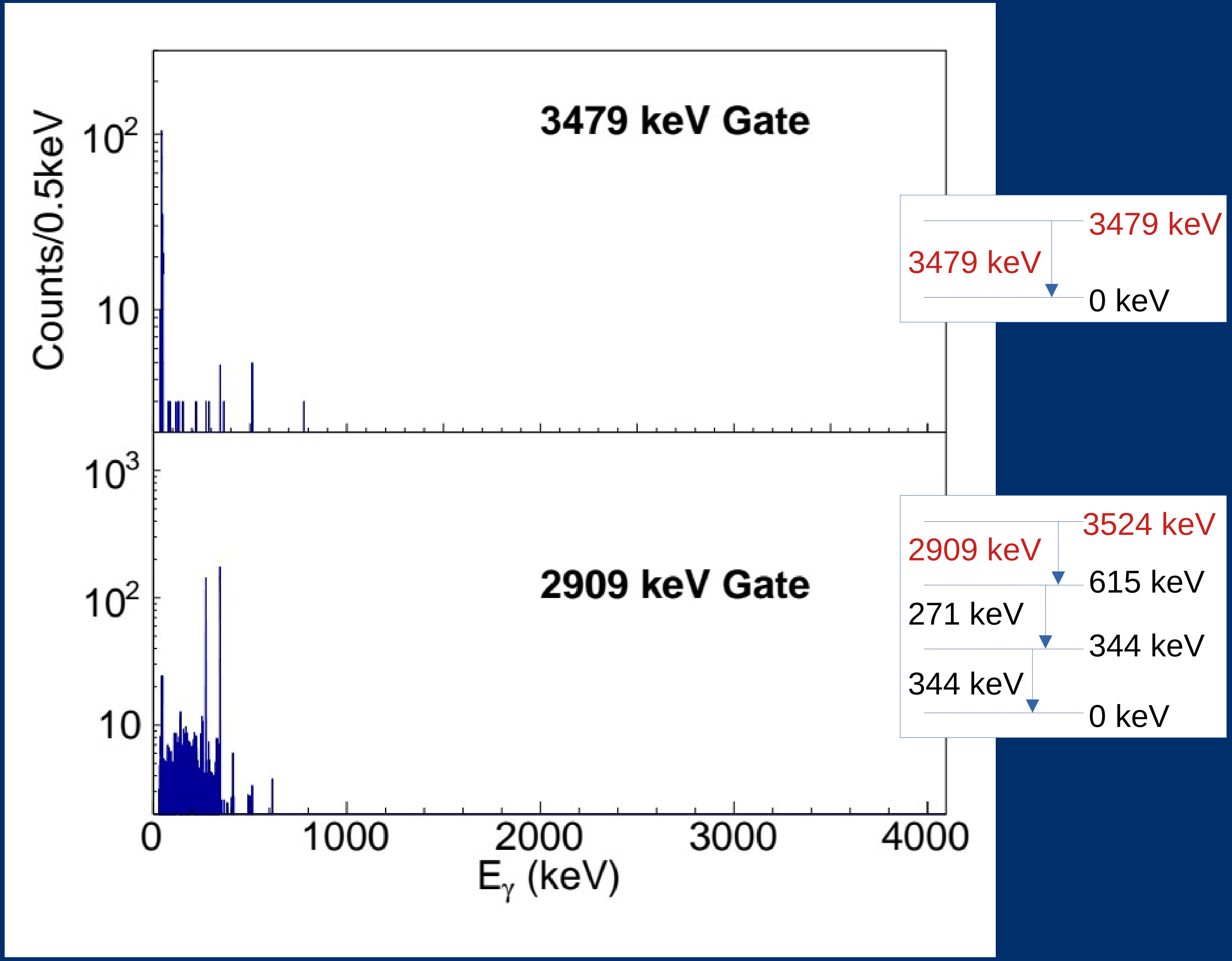
Placing New Transitions

- Verify placement of previously unidentified transitions by **reversing the gating**
- The **entire de-excitation cascade** should appear in the coincidence gate
- Highest energy state previously identified: 3358 keV



Placing New Transitions

- Straight to ground transitions only in coincidence with x-rays
- Cascades may involve intermediate levels



Angular Correlation Analysis

NPL

- Use angular correlations to **assign spin** to previously unidentified levels
- Angular momentum transfer in the decay determines **angular distribution** of emitted gamma rays
- Probe this distribution using coincidences between **different detector pairs**