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

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^{152}Tb: Medical Imaging

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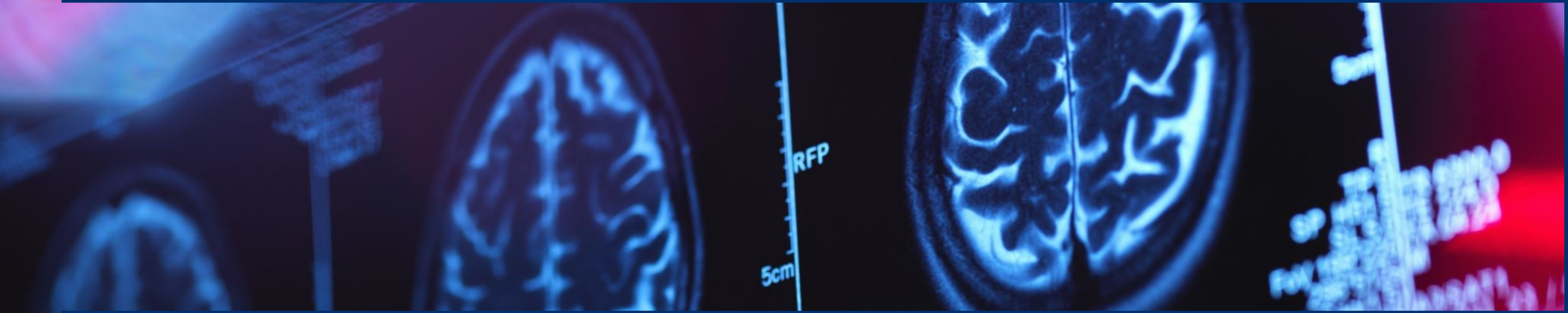
- β^+ / EC decay to ^{152}Gd – **Positron Emitter**
- $T_{1/2}$ = 17.8784(95) h
- Q_{EC} = 3990(40) keV [1]
- Firs-in-human trials show promise in **PET imaging**: ^{152}Tb-DOTATOC and ^{152}Tb-PSMA-617 used successfully in **human patients** [2,3]
- Terbium isotope – applications in **theragnostics**



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

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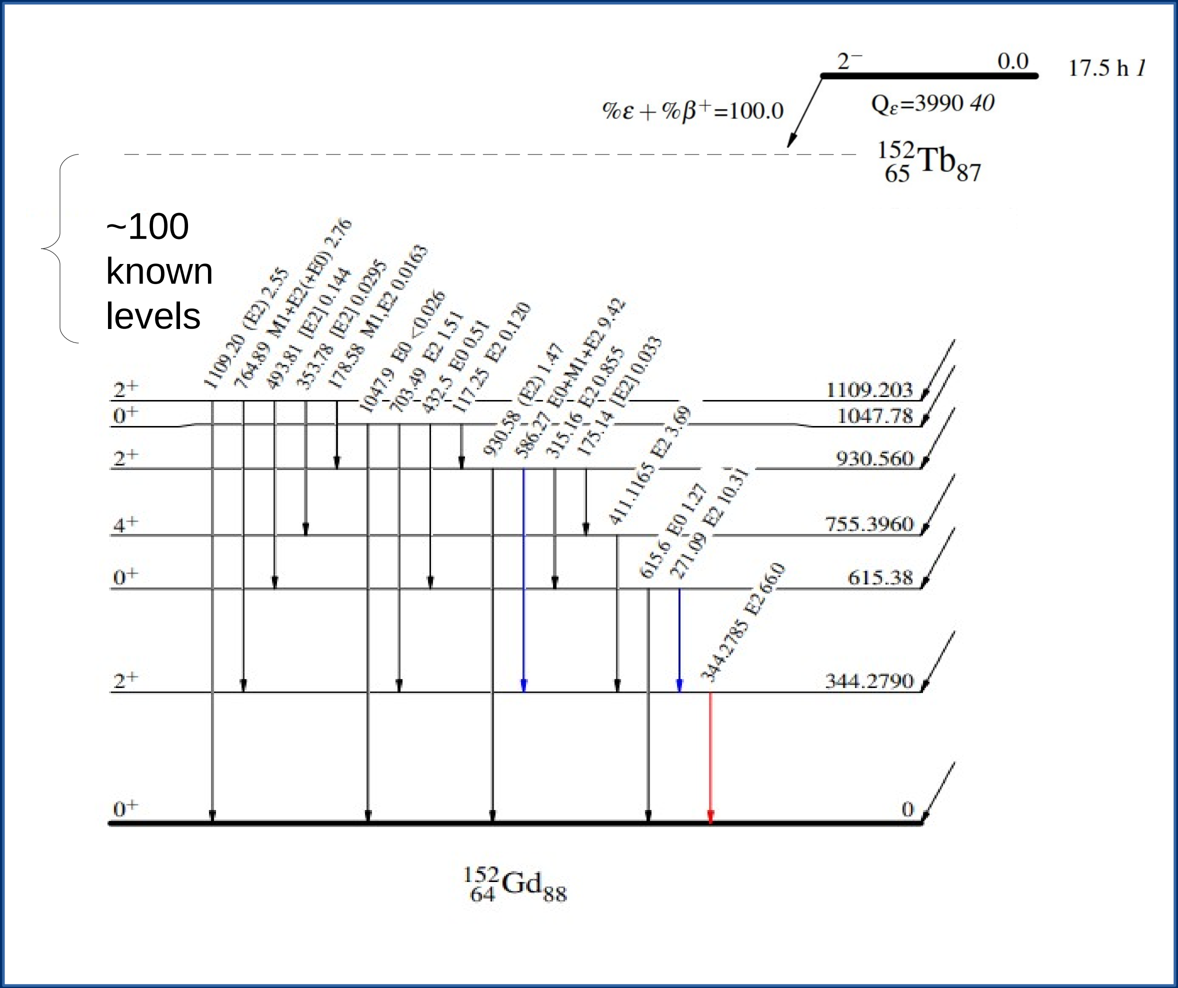
- **Terbium theragnostic quartet**: four different medical uses
- Shared chemistry – compatible with the **same delivery mechanism**
- **Personalised medicine** – treatment plan tailored to individual patients

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

- ^{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**
- 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) State-of-the-art γ-ray assay of ^{86}Y for medical imaging, A.C. Gula et. al.

^{152}Tb Decay Spectroscopy

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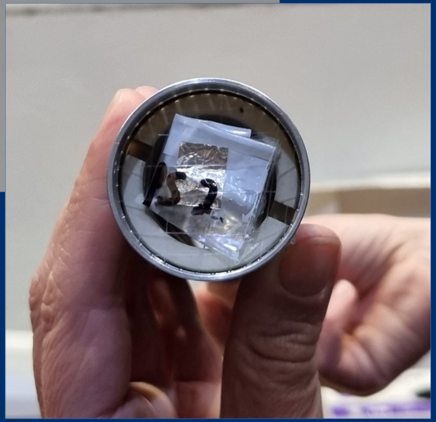
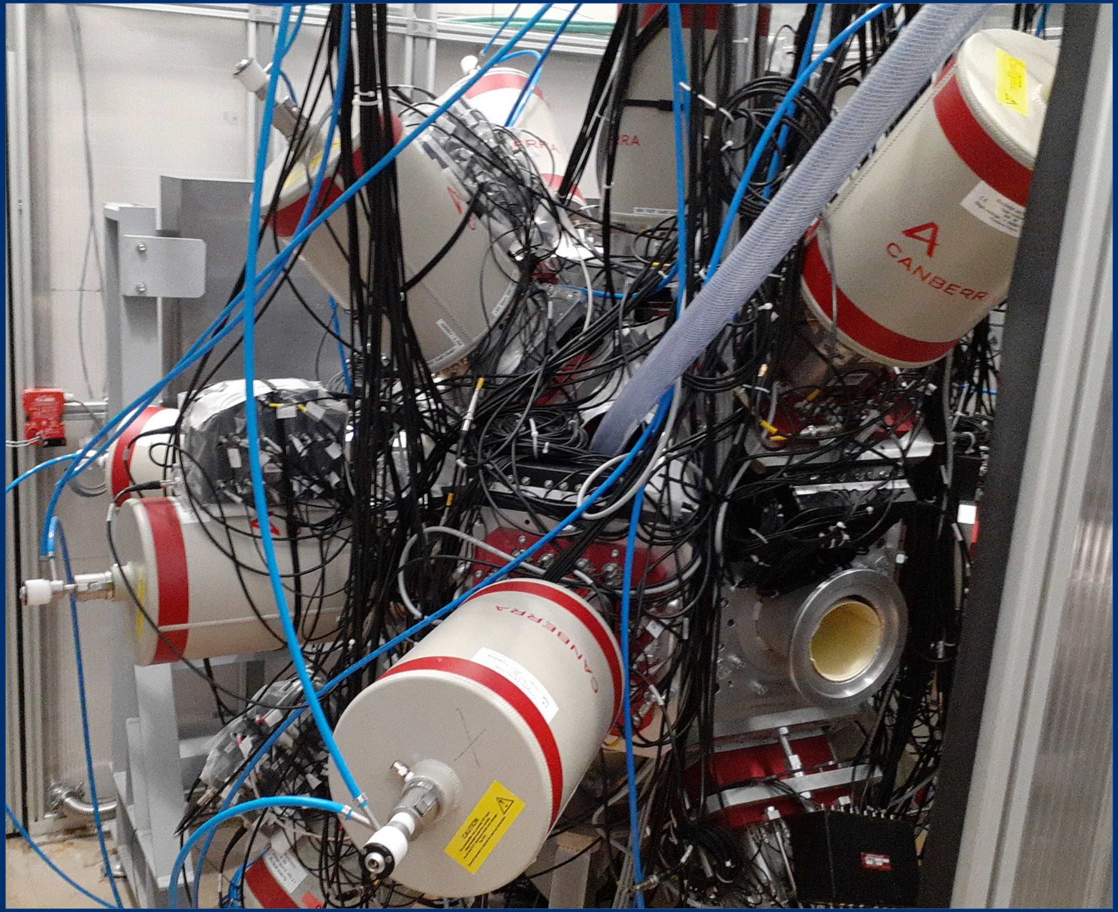
- Sources prepared at CERN ISOLDE: **1.4 GeV proton beam** on a tantalum target
- Samples purified by **laser ionisation** and **mass separation** and implanted onto a pair of Al foils plus one Mylar foil
- Delivered to ILL Grenoble for measurement: **100 and 500 kBq** at start of experiment (3rd May 2023)



Gamma-Gamma Spectroscopy

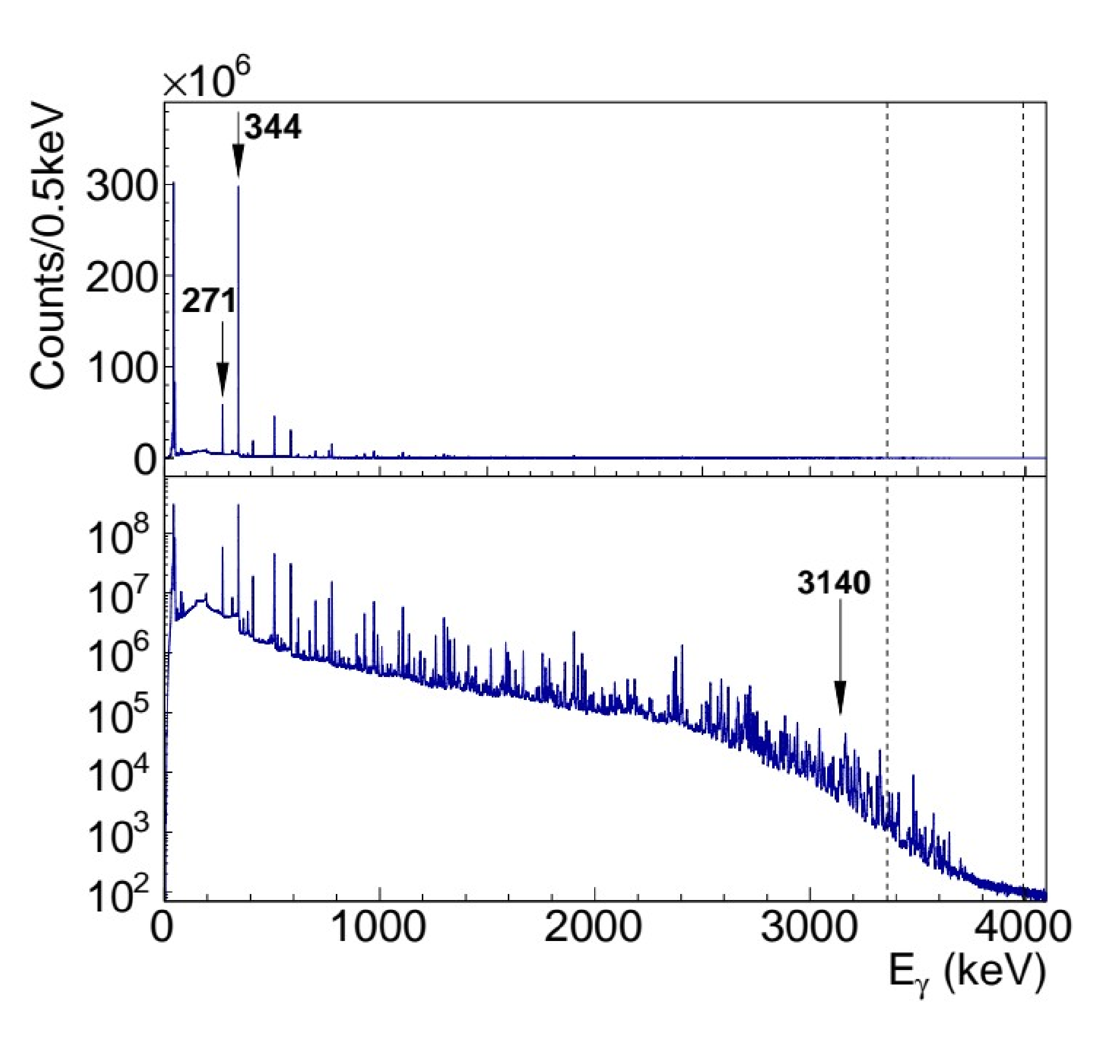
- FIssion Product Prompt Gamma-Ray Spectrometer (FIPPS) [10]
- **64 HPGe crystals**, 16 clovers with BGO shielding (14 crystals excluded)
- Absolute efficiency ~5.6% at 344 keV
- **48 hours** measurement time
- Electron-Gamma spectroscopy carried out in parallel – PN1/LOHENGRIN



10) 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**



Placing New Transitions

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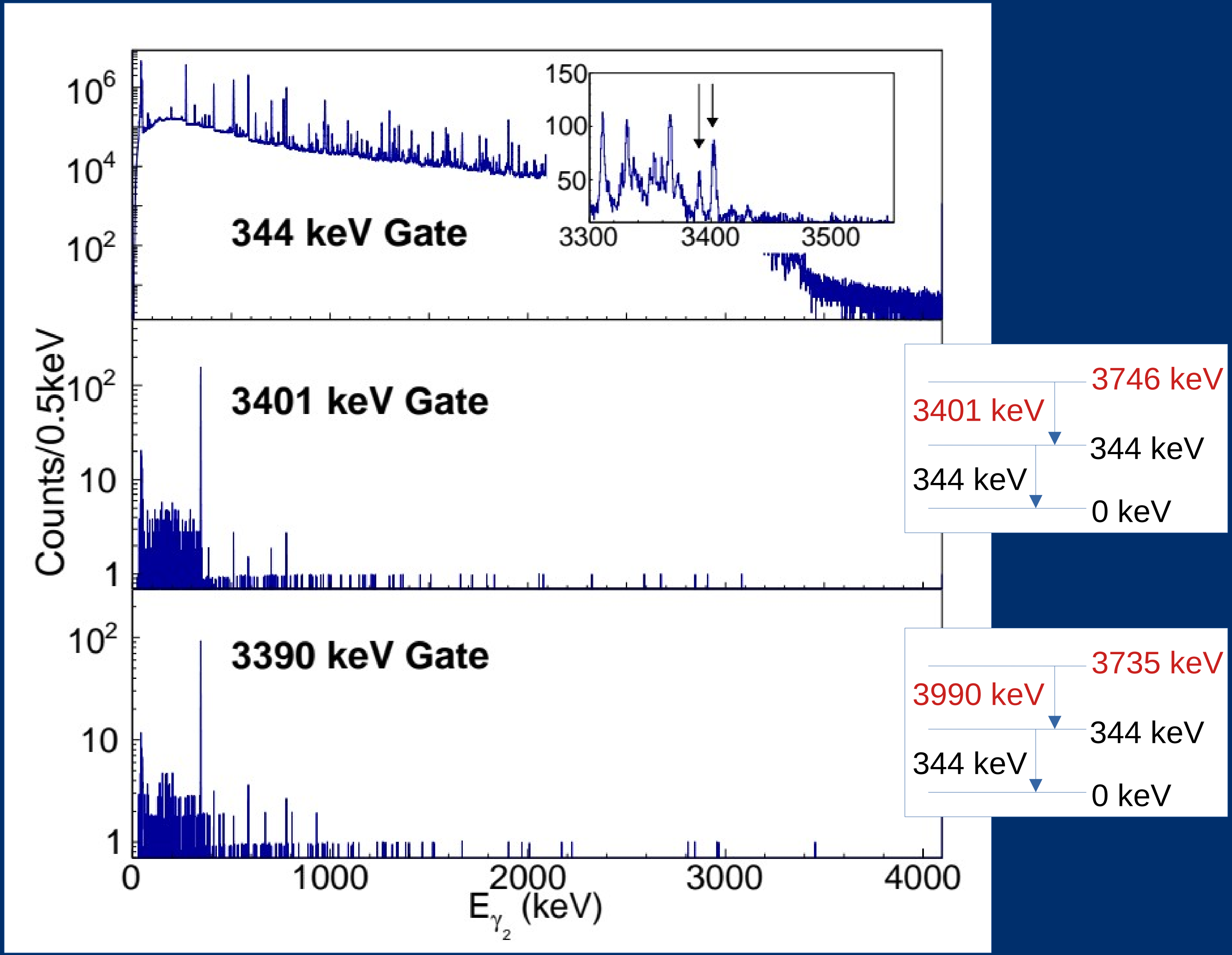
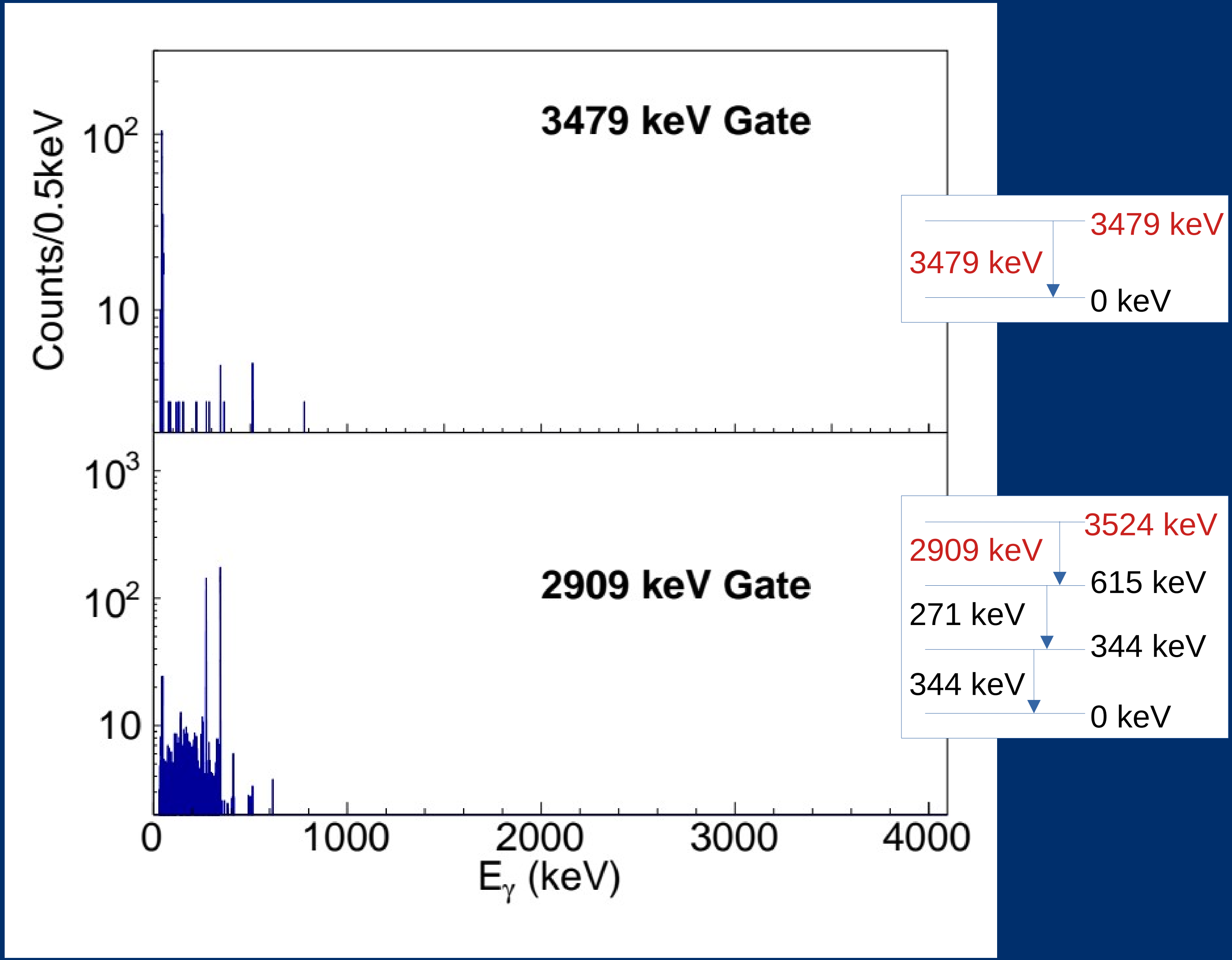
- Verify placement of previously unidentified transitions by **reversing the gating**
- The **entire de-excitation cascade** should appear in the coincidence gate
- Highest energy state previously identified: 3358 keV



Placing New Transitions

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



Angular Correlation Analysis

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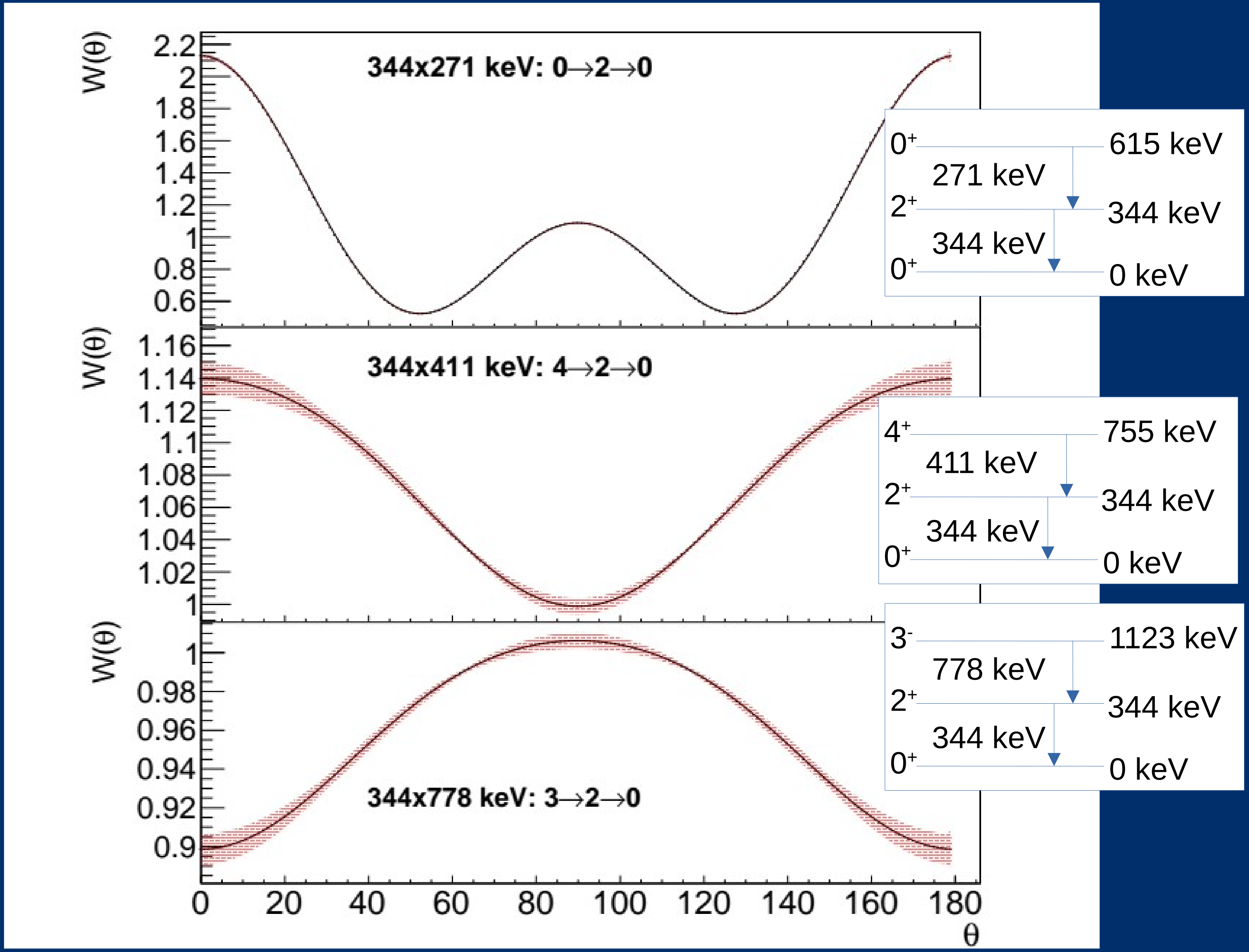
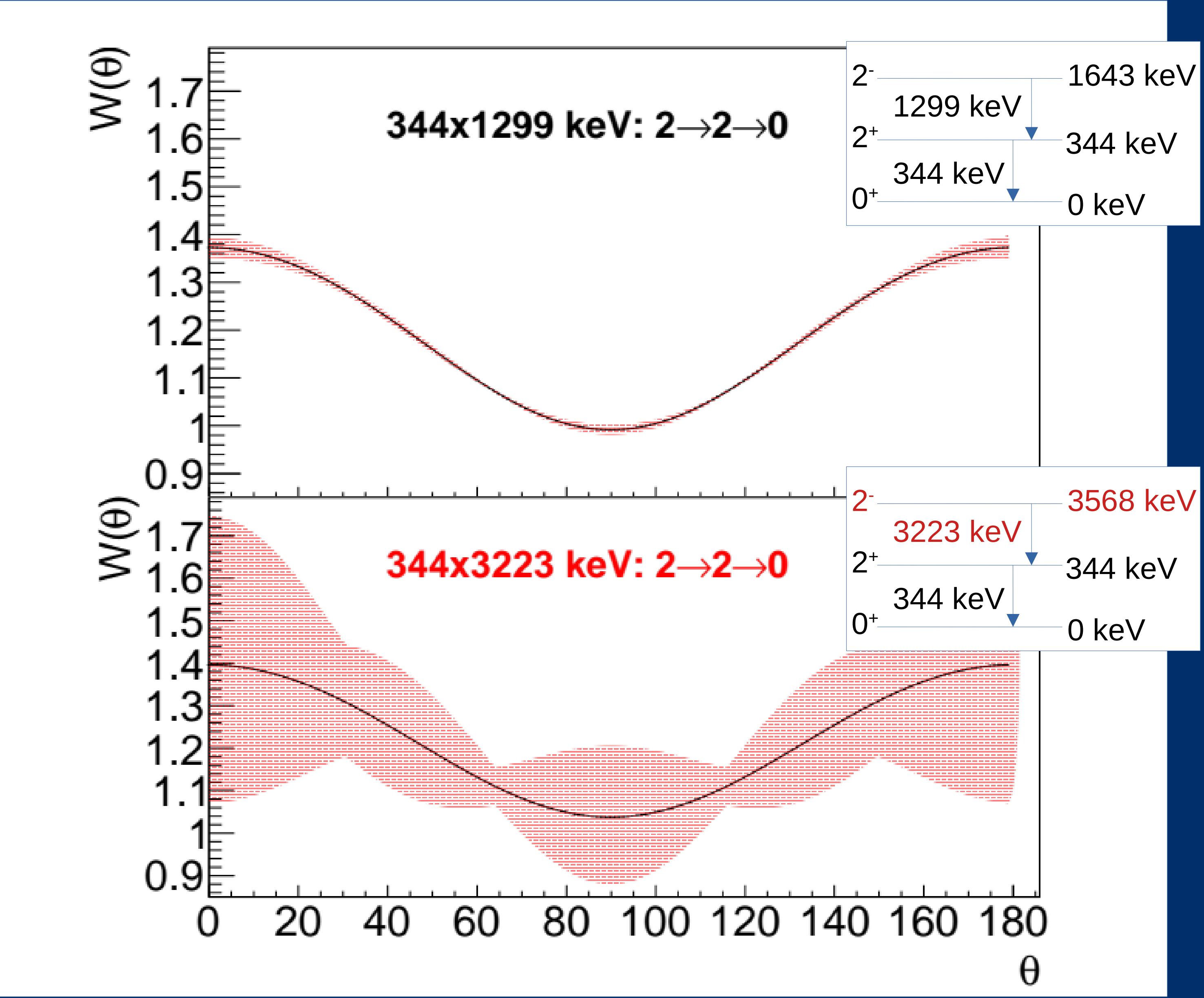
- Use angular correlations to **assign spin** to previously unidentified levels
- Angular momentum transfer in the decay determines **angular distribution** of emitted gamma rays
- Probe this distribution using coincidences between **different detector pairs**



Spin/Parity of New States

- ^{152}Tb ground state has **$J^{\pi}=2^-$**
- "Allowed" beta decays populate **$1^-,2^-,3^-$** in ^{152}Gd
- 2^- to 0^+ ground state **M2**
- 2^+ to 0^+ **E2**
- **No ground state transition** (3568 keV) seen: assign negative parity



Electron-Gamma Spectroscopy

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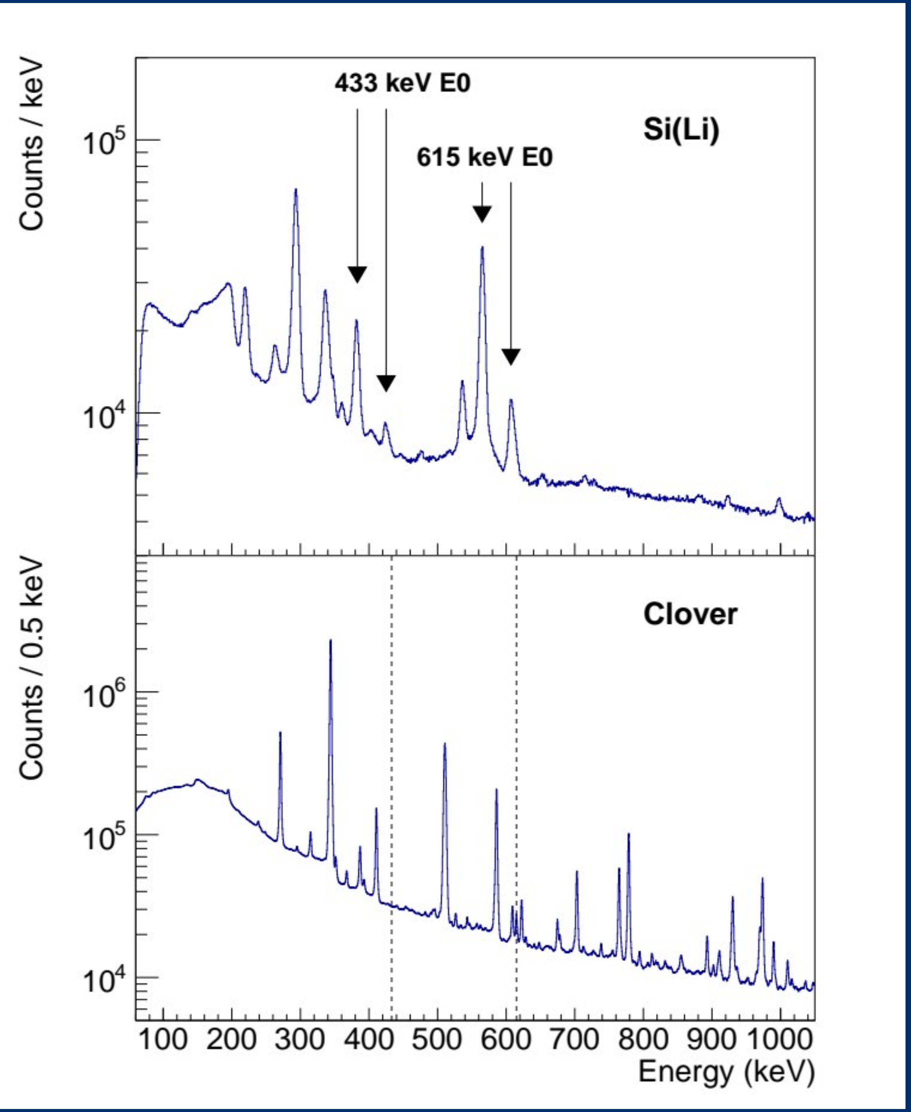
- Internal conversion – electrons emitted instead of gammas
- Invisible to HPGe array
- Electron energy depends on orbital – K & L peaks
- 0+ to 0+ transitions – no single-gamma decay mode, E0 transitions only emit electrons



Preliminary Results + Future Work

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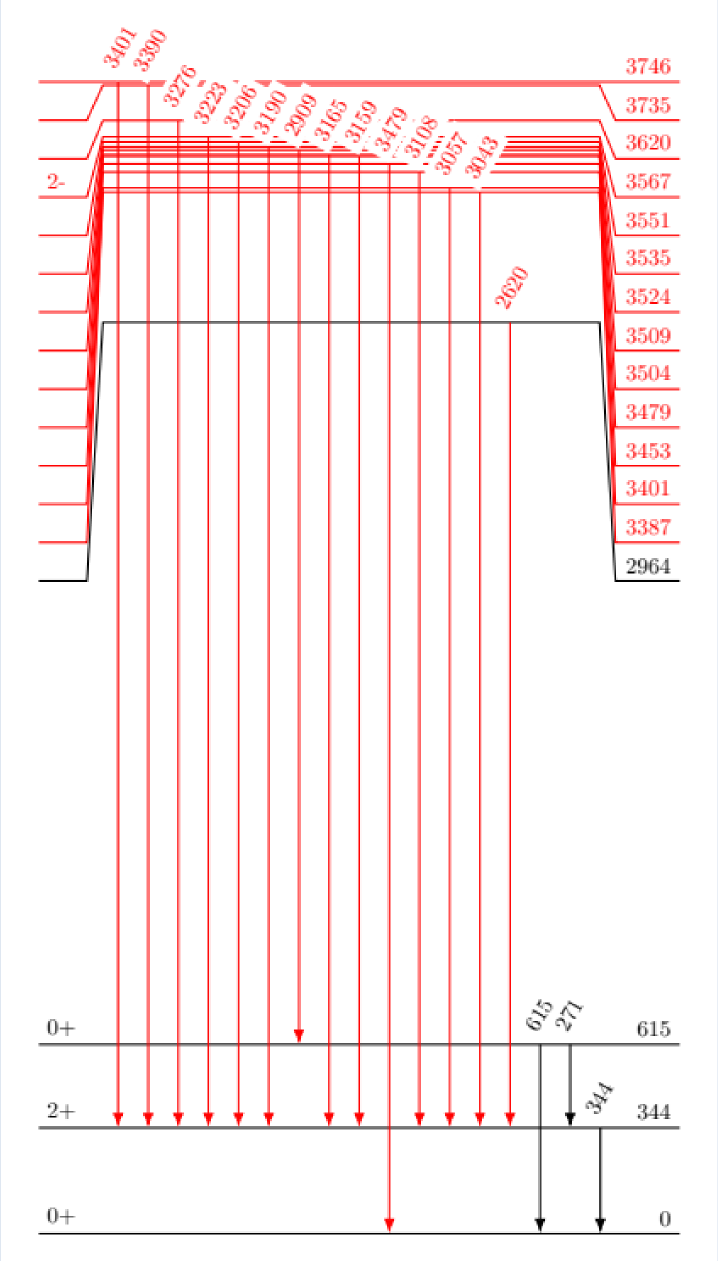
- Over **50 new transitions** placed so far, including to **19 previously unidentified levels**
- Supported by conversion electron spectroscopy – E0 transitions and internal conversion represent **“missing” intensity**
- Transition intensities require **Monte Carlo simulations** to validate efficiency curve
- **Balancing of gamma feeding** in and out of levels to estimate beta feeding intensity
- Calculation of the **overall beta strength** function from the finalised level scheme



11) Electron-Gamma Decay Spectroscopy of 152Tb, E.B. O’Sullivan et al, submitted
12) Towards Complete Decay Spectroscopy of 152Tb, E.B. O’Sullivan et al, submitted

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. State-of-the-art γ-ray assay of ^{86}Y for medical imaging, A.C. Gula et al. Phys. Rev. C 102 (2003) 034316
10. FIPPS (FIssion Product Prompt γ-ray Spectrometer) and its first experimental campaign, C. Michelagnoli et al, EPJ Web Conf. 193, (2018) 04009
11. Electron-gamma spectroscopy of 152Tb, E.B. O’Sullivan et al, Physica Scripta, submitted
12. Towards complete decay spectroscopy of 152Tb, E.B. O’Sullivan et al, Radiation Physics and Chemistry, submitted

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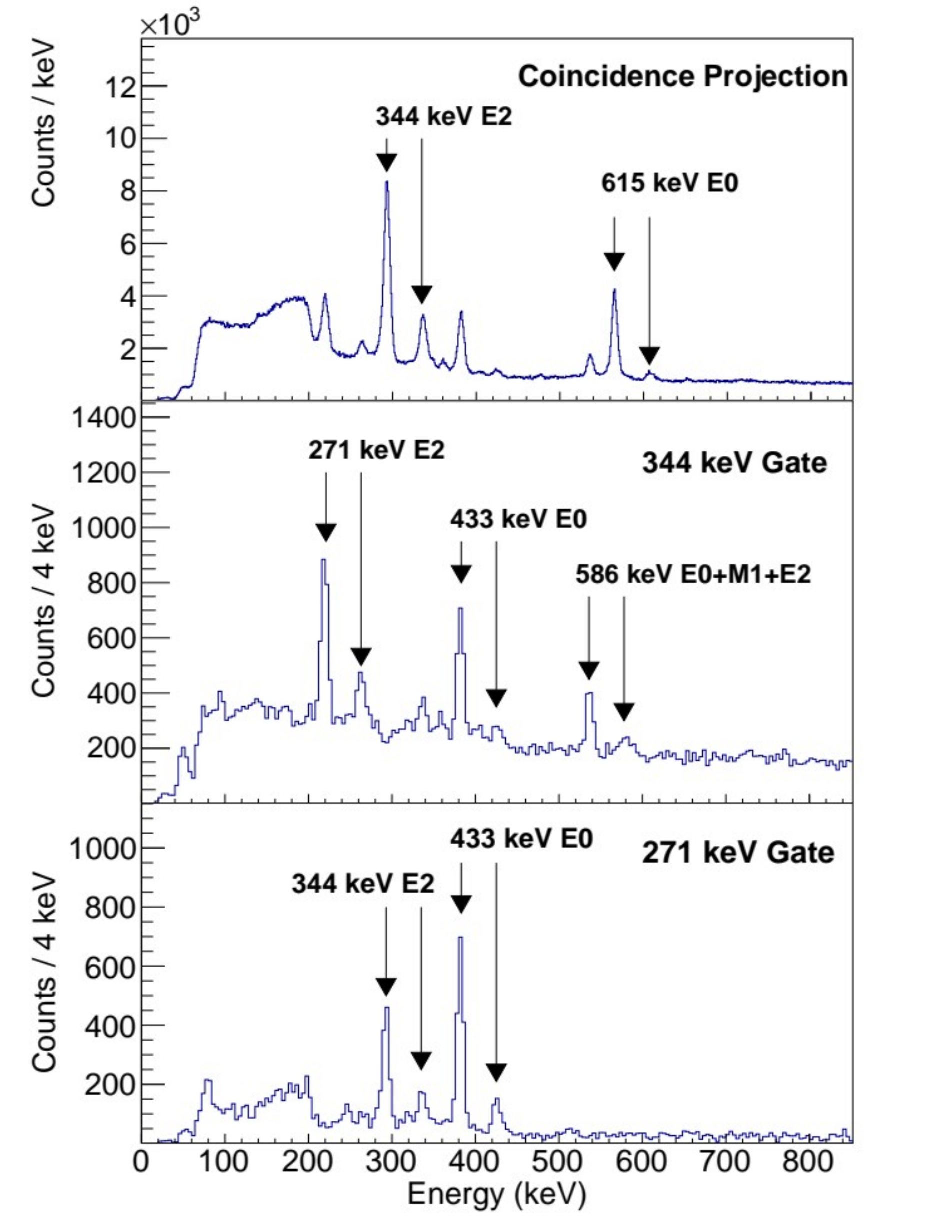
K/L Ratios

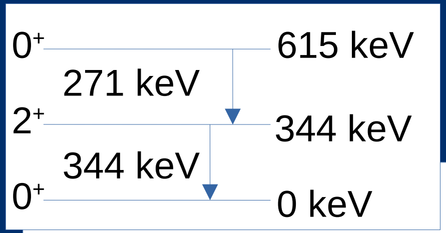
- Use gamma-ray gate on HPGe for selectivity of electron peaks
- K/L ratio depends on transition multipolarity

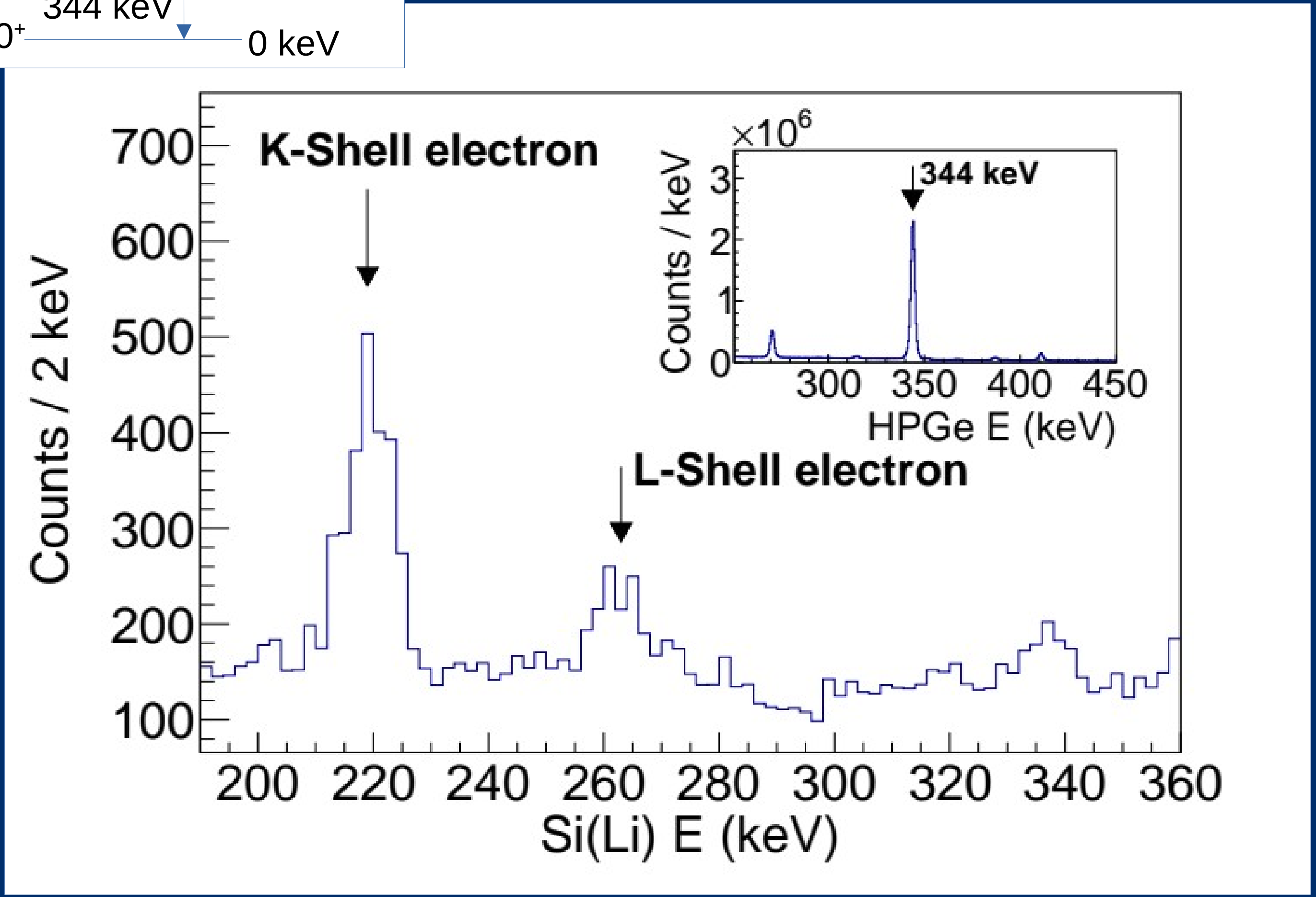
E (keV)	Multipolarity	Measured K/L	Predicted K/L
271	E2	3.8(4)	3.87(8)
344	E2	5.5(7)	4.58(9)
586	E2+M1+E0	4.0(12)	6.13(14)



Internal Conversion

- Gate on gamma peaks to **investigate conversion electrons**
- Further **“missing” intensity** from gamma spectra
- K/L ratio provide **support for multipolarity assignments**
- 271 keV: measured **K/L 4.17**(36), vs BRICC theoretical value of **3.87(8)** for E2 [11]
- Preliminary efficiency correction from GEANT4 simulation of the setup

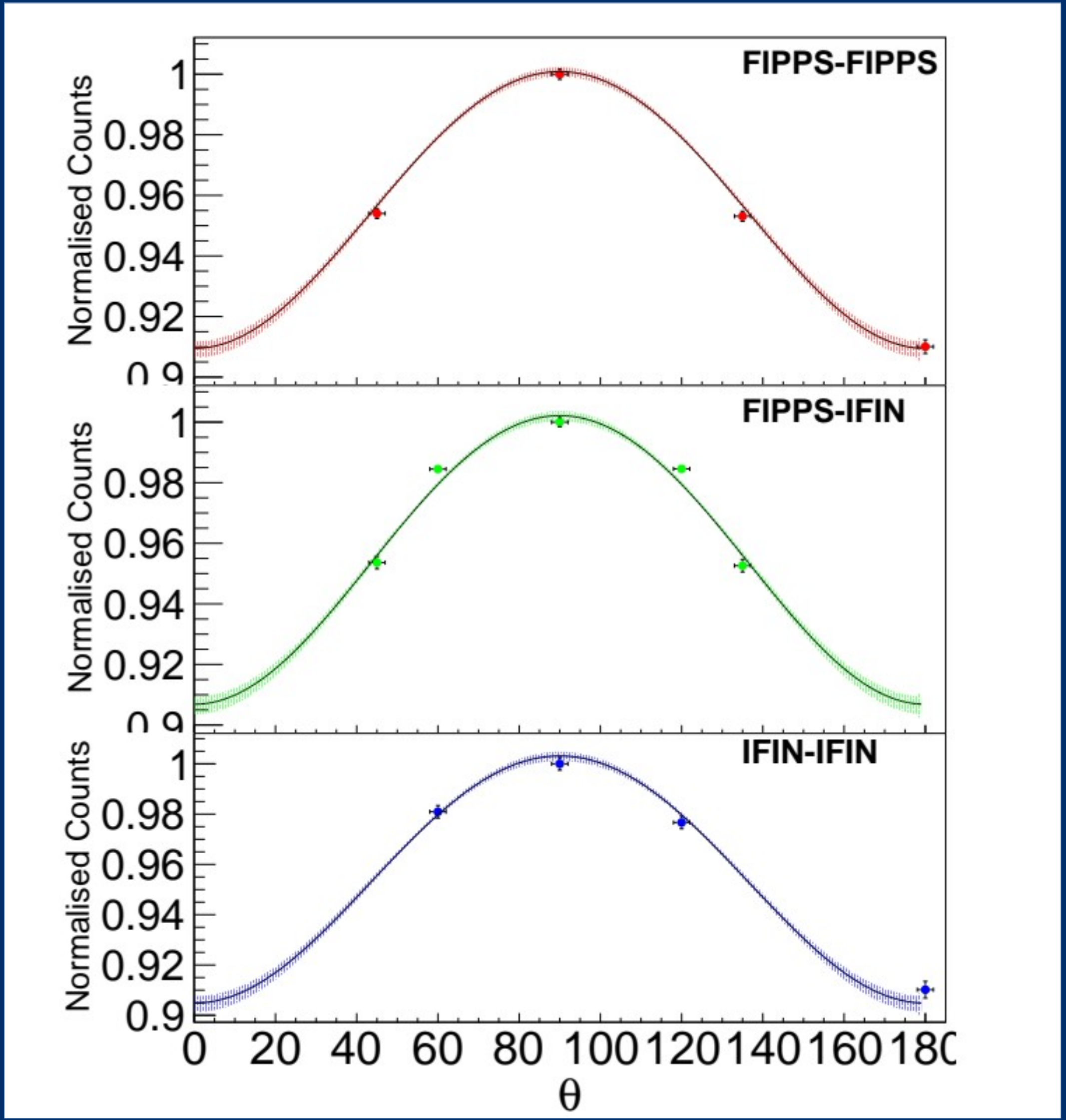




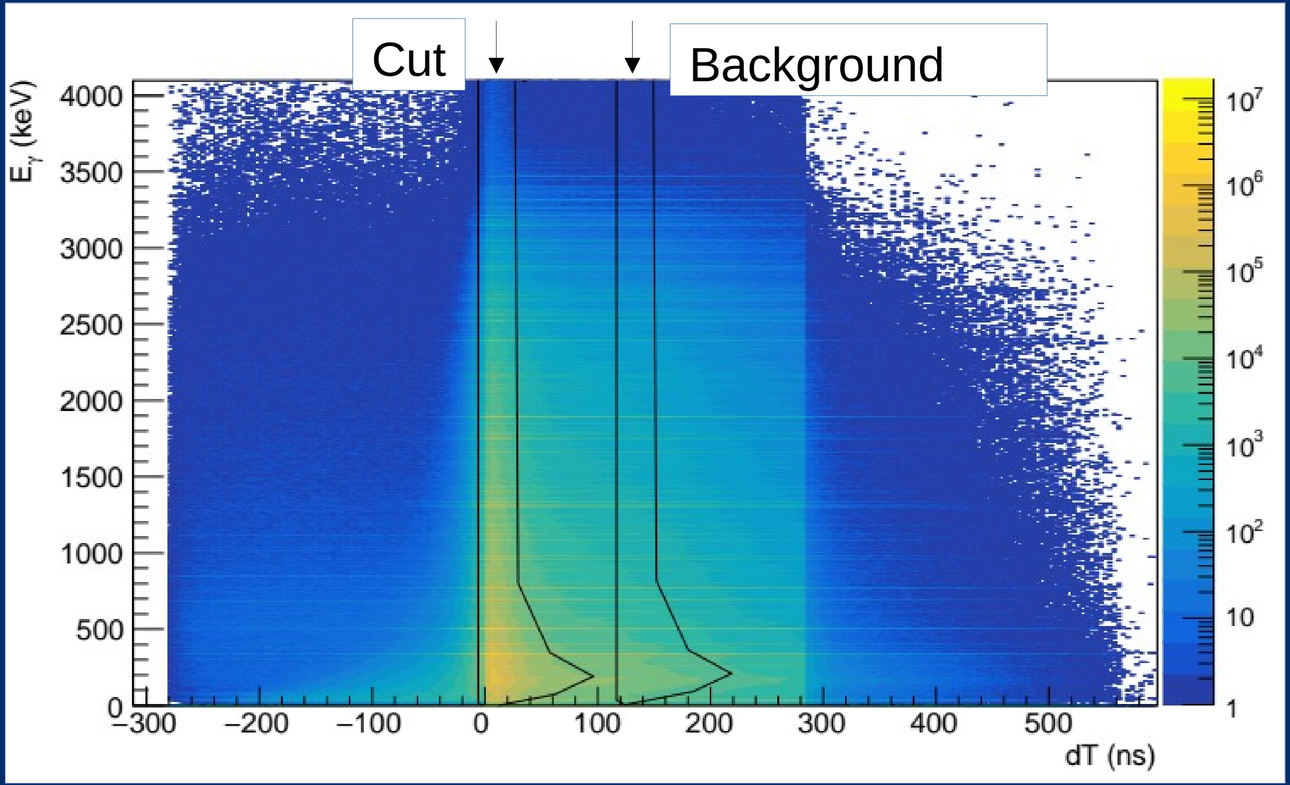
11) Evaluation of theoretical conversion coefficients using BrIcc, T. Kibédi et al
12) Initial Electron-Gamma Spectroscopy of 152Tb, E.B. O’Sullivan et al, in preparation

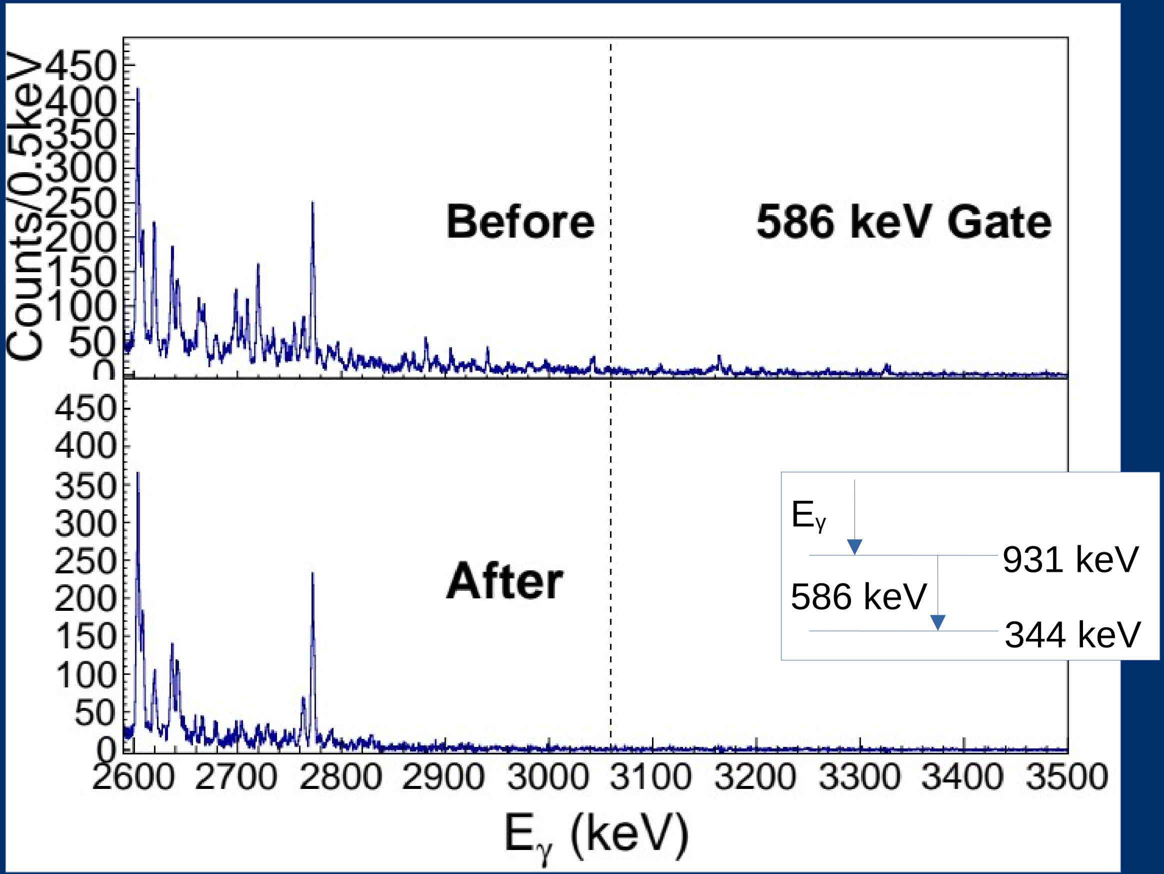
Angular Correlation Minimisation

- Two HPGe detector types – three possible pairings, 6 possible angles
- Fit overall angular correlation function by simultaneously minimising three distributions



Prompt Time Cut





- Select only gammas within 40 ns window – **emitted from same nucleus**
- Offset window 100 ns to **sample background** for subtraction
- **Random coincidences removed**