SENSITIVE DETECTION OF SPECIAL NUCLEAR MATERIALS FOR RPM APPLICATIONS BASED ON GAMMA-FAST NEUTRON COINCIDENCE COUNTING

Débora TROMBETTA[1], Cibi SUNDARAM[1], Kåre Axell[2], and Bo CEDERWALL[1]*

1) Department of Physics , KTH Royal Institute of Technology
2) Swedish Radiation Safety Authority

International Conference on Nuclear Security: Sustaining and Strengthening Efforts
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* cederwall@nuclear.kth.se

KTH Nuclear Physics Group

Mission – Nuclear Science and Technology

“Fundamental” research funded by VR, KAW, GGS
Understanding the strong force as a manifestation in nuclear properties (even after Higgs is only a fraction of hadron and nuclear masses explained)

- What are the limits for the existence of nuclei?
- How do weak binding and extreme proton-neutron asymmetry affect nuclear properties?
- How do collective phenomea and symmetries emerge in complex nuclei from the interactions between the basic constituents?
- What are the origins of the elements?

“Applied” research **funded by SSM, VR, KTH Innovation, Vinnova**

- Develop radiation sensor applications in Medicine and Industry
  - **Nuclear Safeguards and Security**
  - **Nanodosimetry**
  - **Medical Imaging**

Teaching
Courses on Cand., Master & PhD levels on Gen. Physics, Subatomic physics, Experimental techniques in Nuclear and Particle Physics and Radiation protection
Master’s programme in Nuclear Energy Technology

Outreach
Radioactive Orchestra http://www.nuclear.kth.se/radioactiveorchestra/
Berkeley Radwatch project https://radwatch.berkeley.edu/dosenet/map

KTH Nuclear Physics Group

Experiment

Prof. Bo Cederwall (Head of Division)
Prof. Ayse Ataç Nyberg
Dr Torbjörn Bäck, univ.lektor
Prof. em. Arne Johnson
Dr Débora Trombetta, researcher
Dr Biswarup Das, postdoc
Dr Alf Göök, researcher
Özge Aktas, PhD stud.
Linda Eliasson, PhD stud.
Aysegul Ertoprak, PhD stud.
Xiaoyu Liu, PhD stud.
Jana Petrović, PhD stud.
Wei Zhang, PhD stud.
Cibi Sundaram, M.Sc. Stud.
Victor Bussy, M.Sc. Stud.

Kåre Axell, SSM
Tina Sharokhzadeh, SSM

Theory

Assoc. Prof. Chong Qi
Prof. em. Ramon Wyss
Prof. em. Roberto Liotta
Daniel Karlsson, PhD stud.
Karl Sallmén, M.Sc. stud.
Akshay Kishore Kallianpur, M.Sc. Stud.

KTH Nuclear Safeguards and Security

Development of radiation detection and imaging systems for applications in nuclear safeguards and non-proliferation, nuclear security, environment and related areas

SSM competence centre for radiation detection

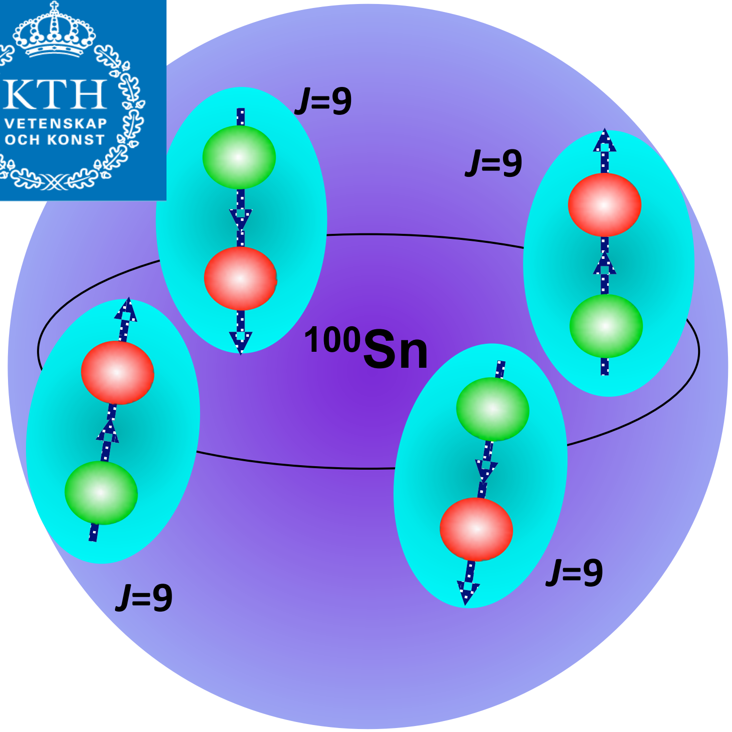
External funding: Swedish Radiation Safety Autority (SSM)
Swedish Research Council (VR)
Swedish Agency for Innovation (Vinnova)

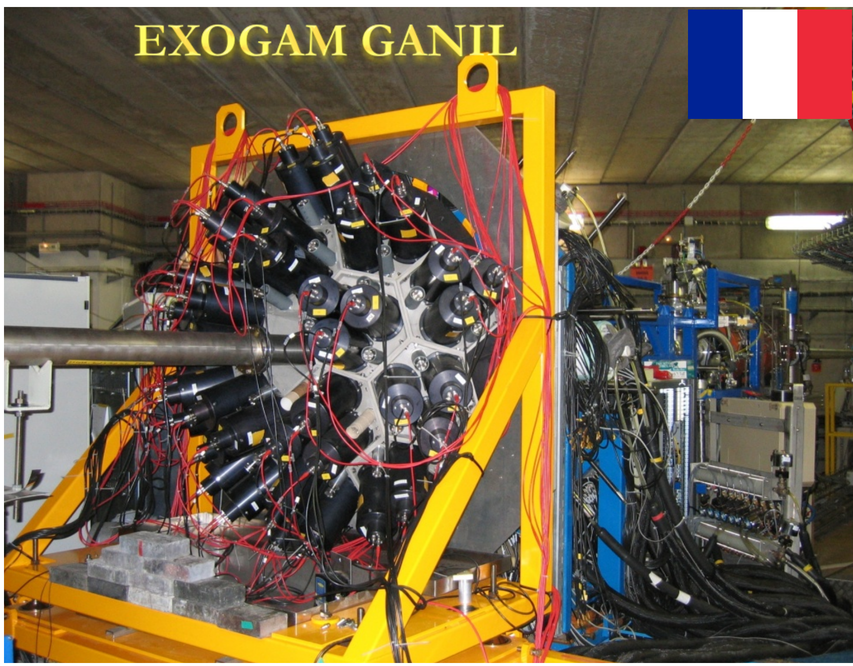
Local team

Bo Cederwall, Prof. Dr.
Débora Trombetta, Dr. (VR Starting grant 2020-2023)
Alf Göök, Dr. (funded by SSM)
Jana Petrović, PhD stud. (funded by SSM)
Cibi Sundaram, M.Sc. Stud.
Victor Bussy, M.Sc. Stud.

External

Kåre Axell, Dr., SSM
Tina Shahrokhzadeh, SSM





LETTER

doi:10.1038/nature09644

Evidence for a spin-aligned neutron–proton paired phase from the level structure of ^{92}Pd

B. Cederwall[1], F. Ghazi Moradi[1], T. Bäck[1], A. Johnson[1], J. Blomqvist[1], E. Clément[2], G. de France[2], R. Wadsworth[3], K. Andgren[1], K. Lagergren[1,4], A. Dijon[2], G. Jaworski[5,6], R. Liotta[1], C. Qi[1], B. M. Nyakó[7], J. Nyberg[8], M. Palacz[5], H. Al-Azri[3], A. Algora[9], G. de Angelis[10], A. Ataç[11], S. Bhattacharyya[2]†, T. Brock[3], J. R. Brown[3], P. Davies[3], A. Di Nitto[12], Zs. Dombrádi[7], A. Gadea[9], J. Gál[7], B. Hadinia[1], F. Johnston-Theasby[3], P. Joshi[3], K. Juhász[13], R. Julin[14], A. Jungclaus[15], G. Kalinka[7], S. O. Kara[11], A. Khaplanov[1], J. Kownacki[5], G. La Rana[12], S. M. Lenzi[16], J. Molnár[7], R. Moro[12], D. R. Napoli[10], B. S. Nara Singh[3], A. Persson[1], F. Recchia[16], M. Sandzelius[1]†, J.-N. Scheurer[17], G. Sletten[18], D. Sohler[7], P.-A. Söderström[8], M. J. Taylor[3], J. Timár[7], J. J. Valiente-Dobón[10], E. Vardaci[12] & S. Williams[19]

Shell structure and magic numbers in atomic nuclei were generally explained by pioneering work[1] that introduced a strong spin–orbit interaction to the nuclear shell model potential. However, knowledge of nuclear forces and the mechanisms governing the structure of nuclei, in particular far from stability, is still incomplete. In nuclei with equal neutron and proton numbers ($N = Z$), enhanced correlations arise between neutrons and protons (two distinct types of fermions) that occupy orbitals with the same quantum numbers. Such correlations have been predicted to favou
unusual type of nuclear superfluidity, termed isoscalar
proton pairing[2–6], in addition to normal isovector
many experimental efforts, these predictio
firmed. Here we report the experim
states in the $N = Z = 46$ nucleus ^{92}
ing the ^{58}Ni(^{36}Ar,2n)^{92}Pd
tified using a com
γ-ray, charged
reveal ex
ggest
ty (charac-
d and low-lying
Such strong, isoscalar
ave a considerable impact on
possibly influence the dynamics of
tellar nucleosynthesis.

clei, including those residing along the $N = Z$ line
mass 80, a detailed analysis of properties such as binding
and the spectroscopy of excited states[10] strongly suggests that
ormal isovector (isospin $T = 1$, see Fig. 1) pairing is dominant at low excitation energies. On the other hand, there are long-standing predictions[2–6] for a change in the heavier $N = Z$ nuclei, from a nuclear superfluid dominated by isovector pairing to a structure where isoscalar ($T = 0$) neutron–proton (np) pairing has a major influence, as the mass number increases towards the exotic doubly magic nucleus ^{100}Sn, the heaviest $N = Z$ nucleus predicted to be bound.

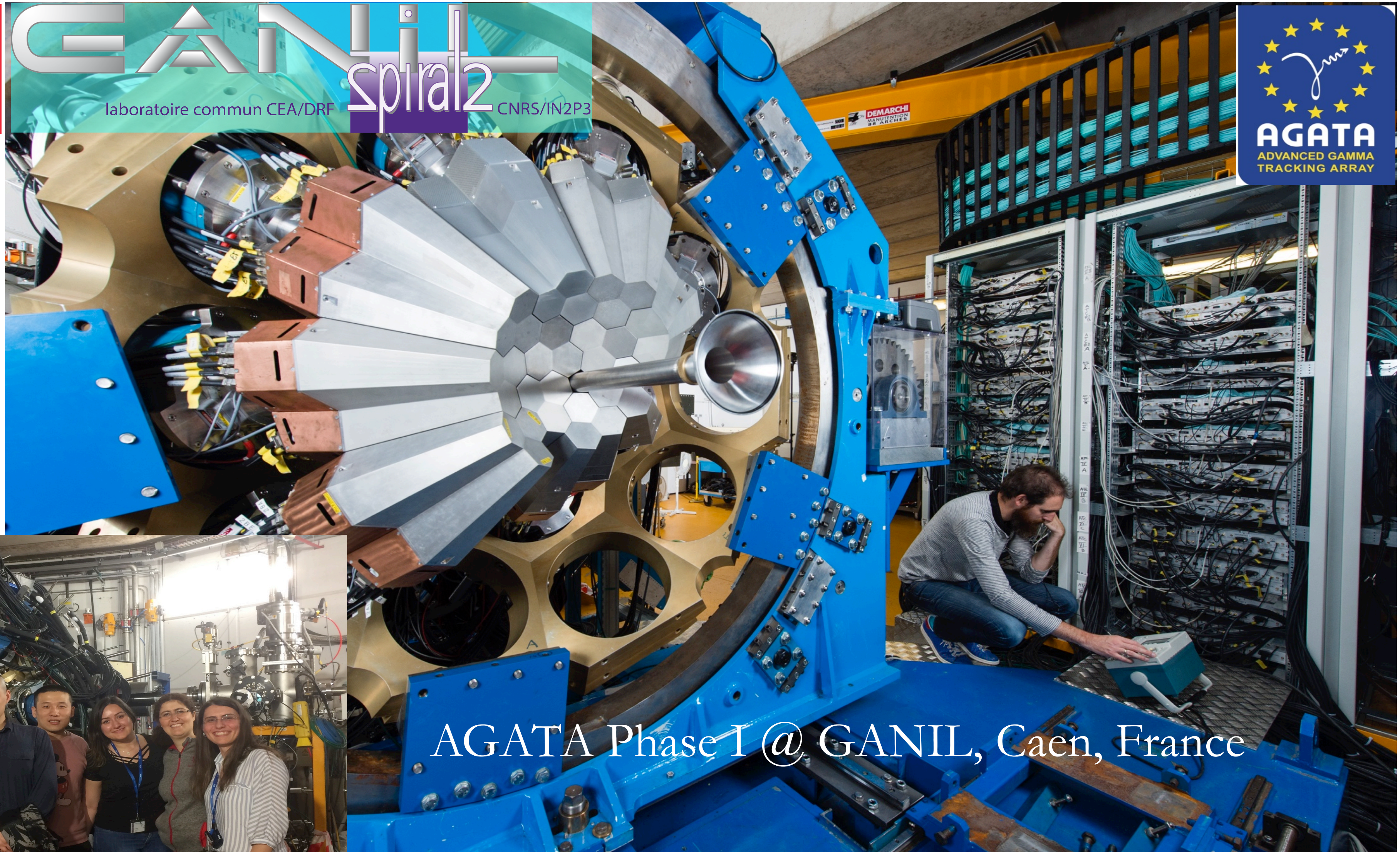
Nuclei with $N = Z$ and mass number >90 can only be produced in the laboratory with very low cross-sections. The related problems of identifying and distinguishing such reaction products and their associated γ-rays from the vast array of $N > Z$ nuclei
greater numbers from the reactions
of their low-lying excited state
experimental difficulties
efficient detector sy

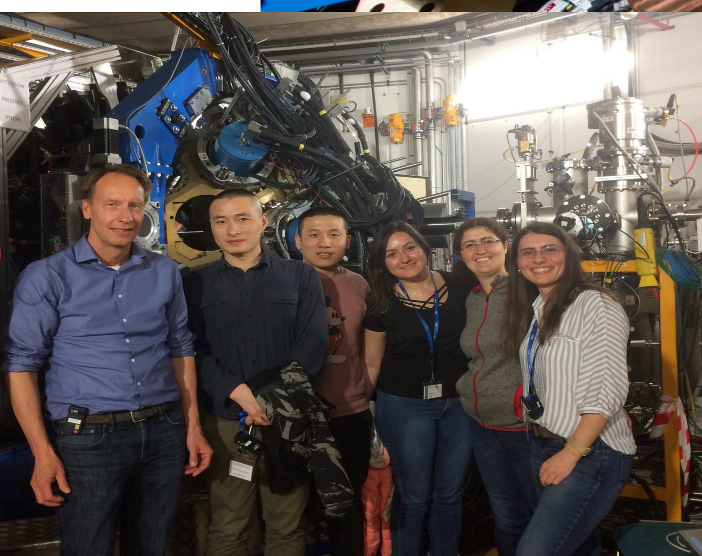
Excited st
evapo
ns
MeV,
%) ^{58}Ni target.
ticles), neutrons and
in coincidence. A schem
p is shown in Fig. 2.
aporation reaction channel following
compound nucleus, leading to ^{92}Pd, was very
, with a relative yield of less than 10^{-5} of the total
-section. Gamma-rays from decays of excited states in ^{92}Pd
identified by comparing γ-ray spectra in coincidence with two emitted neutrons and no charged particles with γ-ray spectra in coincidence with other combinations of neutrons and charged particles. The typical efficiency for detecting any charged particle was 66%. This number rises to 88% or higher if more than one such particle is emitted in a particular reaction channel. The clean identification of neutrons is crucial, as scattering of neutrons from one detector segment to another can be misinterpreted as two neutrons; this would give rise to a background from the much more prolific reaction channels (where only one neutron has been emitted) in γ-ray spectra gated by two neutrons. But because neutrons have a finite velocity, the difference in detection

Figure 1 | Schematic illustration of the two possible pairing schemes in nuclei. a, The normal isospin $T = 1$ triplet. The two like-particle pairing components are responsible for most known effects of nuclear superfluidity. Within a given shell these isovector components are restricted to spin zero owing to the Pauli principle. **b**, Isoscalar $T = 0$ neutron–proton pairing. Here the Pauli principle allows only non-zero components of angular momentum.

[1]Department of Physics, Royal Institute of Technology, SE-10691 Stockholm, Sweden. [2]Grand Accélérateur National d'Ions Lourds (GANIL), CEA/DSM – CNRS/IN2P3, F-14076 Caen Cedex 5, France. [3]Department of Physics, University of York, York YO10 5DD, UK. [4]Joint Institute for Heavy-Ion Research, Holifield Radioactive Ion Beam Facility, Oak Ridge, Tennessee 37831, USA. [5]Heavy Ion Laboratory, University of Warsaw, 02-093 Warsaw, Poland. [6]Faculty of Physics, Warsaw University of Technology, Koszykowa 75, 00-662 Warsaw, Poland. [7]Institute of Nuclear Research of the Hungarian Academy of Sciences, ATOMKI, H-4001 Debrecen, Hungary. [8]Department of Physics and Astronomy, Uppsala University, SE-75121 Uppsala, Sweden. [9]IFIC, CSIC, University of Valencia, E-46071 Valencia, Spain. [10]Instituto Nazionale di Fisica Nucleare, Laboratori Nazionali di Legnaro, I-35020 Legnaro, Italy. [11]Department of Physics, Ankara University, 06100 Tandogan Ankara, Turkey. [12]Dipartimento di Scienze Fisiche, Università di Napoli and Instituto Nazionale di Fisica Nucleare, I-80126 Napoli, Italy. [13]Department of Information Technology, University of Debrecen, H-4010 Debrecen, Hungary. [14]Department of Physics, University of Jyväskylä, FIN-40014 Jyväskylä, Finland. [15]Instituto de Estructura de la Materia, CSIC, E-28006 Madrid, Spain. [16]Diparimento di Fisica dell'Università di Padova and Instituto Nazionale di Fisica Nucleare, Sezione di Padova, I-35122 Padova, Italy. [17]Université Bordeaux 1, CNRS/IN2P3, Centre d'Etudes Nucléaires de Bordeaux Gradignan, F-33175 Gradignan, France. [18]The Niels Bohr Institute, University of Copenhagen, 2100 Copenhagen, Denmark. [19]TRIUMF, Vancouver, British Columbia V6T 2A3, Canada. †Present addresses: VECC, 1/AF Bidhan Nagar, Kolkata 700064, India (S.B.); Department of Physics, University of Jyväskylä, FIN-40014 Jyväskylä, Finland (M.S.).

Fast γ-neutron coincidence detection adapted from fundamental nuclear physics experiments





KTH VETENSKAP OCH KONST

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Accepted Paper

Isospin properties of nuclear pair correlations from the level structure of the self-conjugate nucleus ^{88}Ru

Phys. Rev. Lett.

B. Cederwall et al.

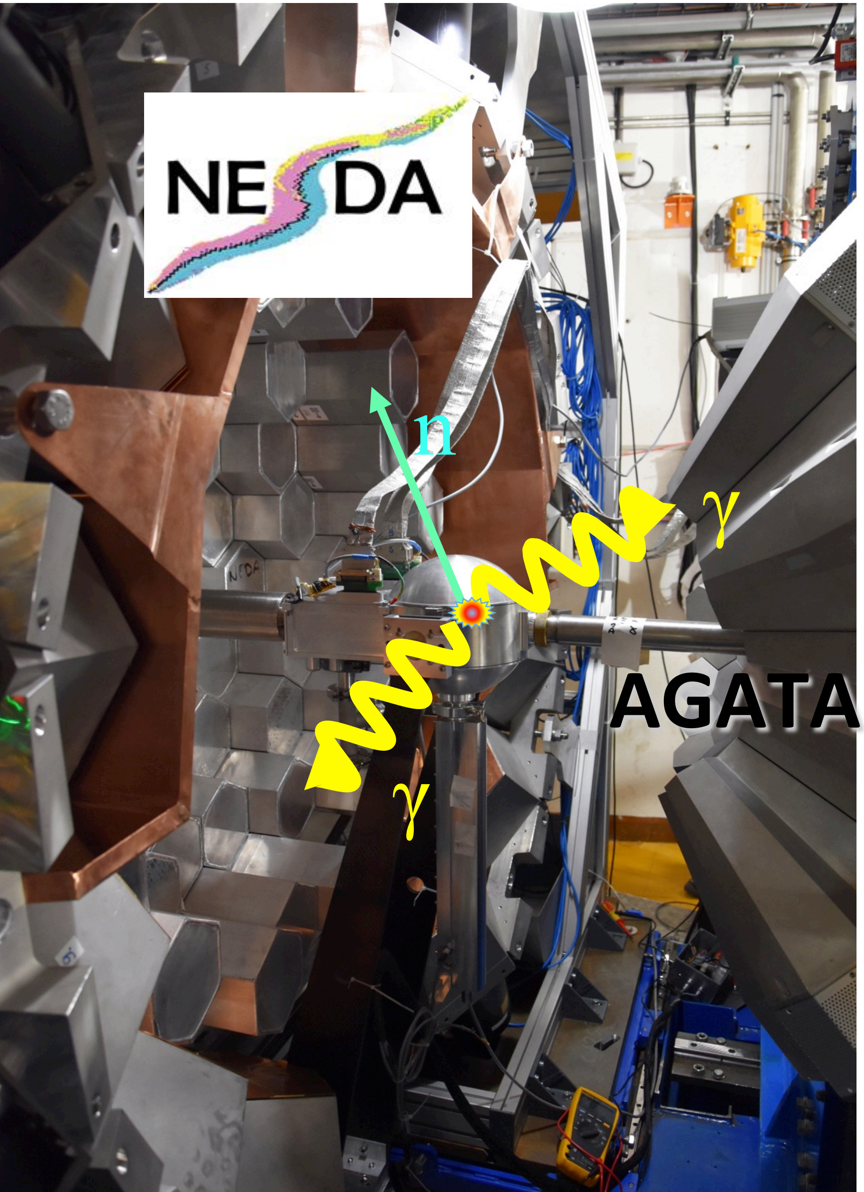
Accepted 18 December 2019

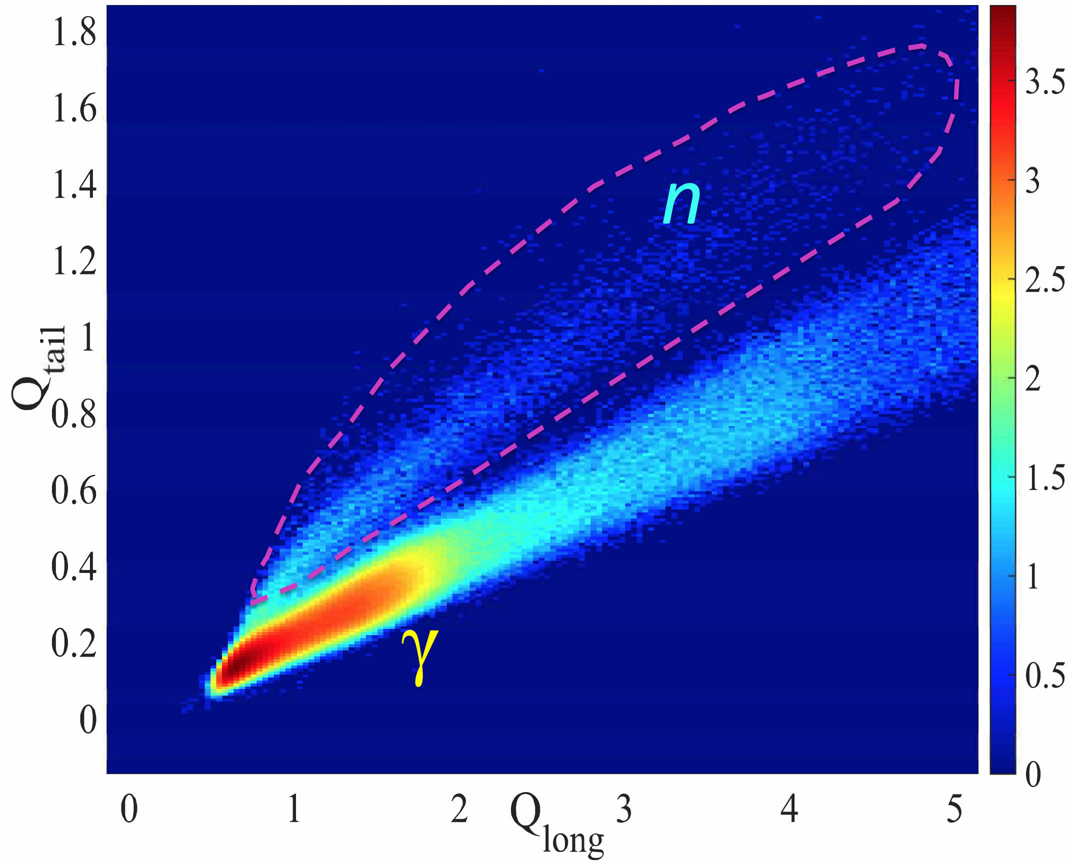
ABSTRACT

ABSTRACT

The low-lying energy spectrum of the extremely neutron-deficient self-conjugate ($N = Z$) nuclide $^{88}_{44}Ru_{44}$ has been measured using the combination of the Advanced Gamma Tracking Array (AGATA) spectrometer, the NEDA and Neutron Wall neutron detector arrays, and the DIAMANT charged particle detector array. Excited states in ^{88}Ru were populated via the $^{54}Fe(^{36}Ar, 2n\gamma)^{88}Ru^{*}$ fusion-evaporation reaction at the Grand Acc'el'erateur National d'Ions Lourds (GANIL) accelerator complex. The observed γ-ray cascade is assigned to ^{88}Ru using clean prompt γ-γ-2-neutron coincidences in anti-coincidence with the detection of charged particles, confirming and extending the previously assigned sequence of low-lying excited states. It is consistent with a moderately deformed rotating system exhibiting a band crossing at a rotational frequency that is significantly higher than standard theoretical predictions with isovector pairing, as well as observations in neighboring $N > Z$ nuclides. The direct observation of such a "delayed" rotational alignment in a deformed $N = Z$ nucleus is in agreement with theoretical predictions related to the presence of strong isoscalar neutron-proton pair correlations.

Pulse shape analysis and time-of-flight measurements for fast neutron-gamma discrimination using organic scintillators





IAEA 2020-02/BC

Fast neutron-gamma correlations for sensitive detection of SNM

Nuclear Inst. and Methods in Physics Research, A 927 (2019) 119–124

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Fast neutron- and γ-ray coincidence detection for nuclear security and safeguards applications

Débora M. Trombetta [a], Malin Klintefjord [a], Kåre Axell [b], Bo Cederwall [a,*]

[a] Department of Physics, KTH Royal Institute of Technology, S-10691 Stockholm, Sweden
[b] Swedish Radiation Safety Authority, S-171 16 Stockholm, Sweden

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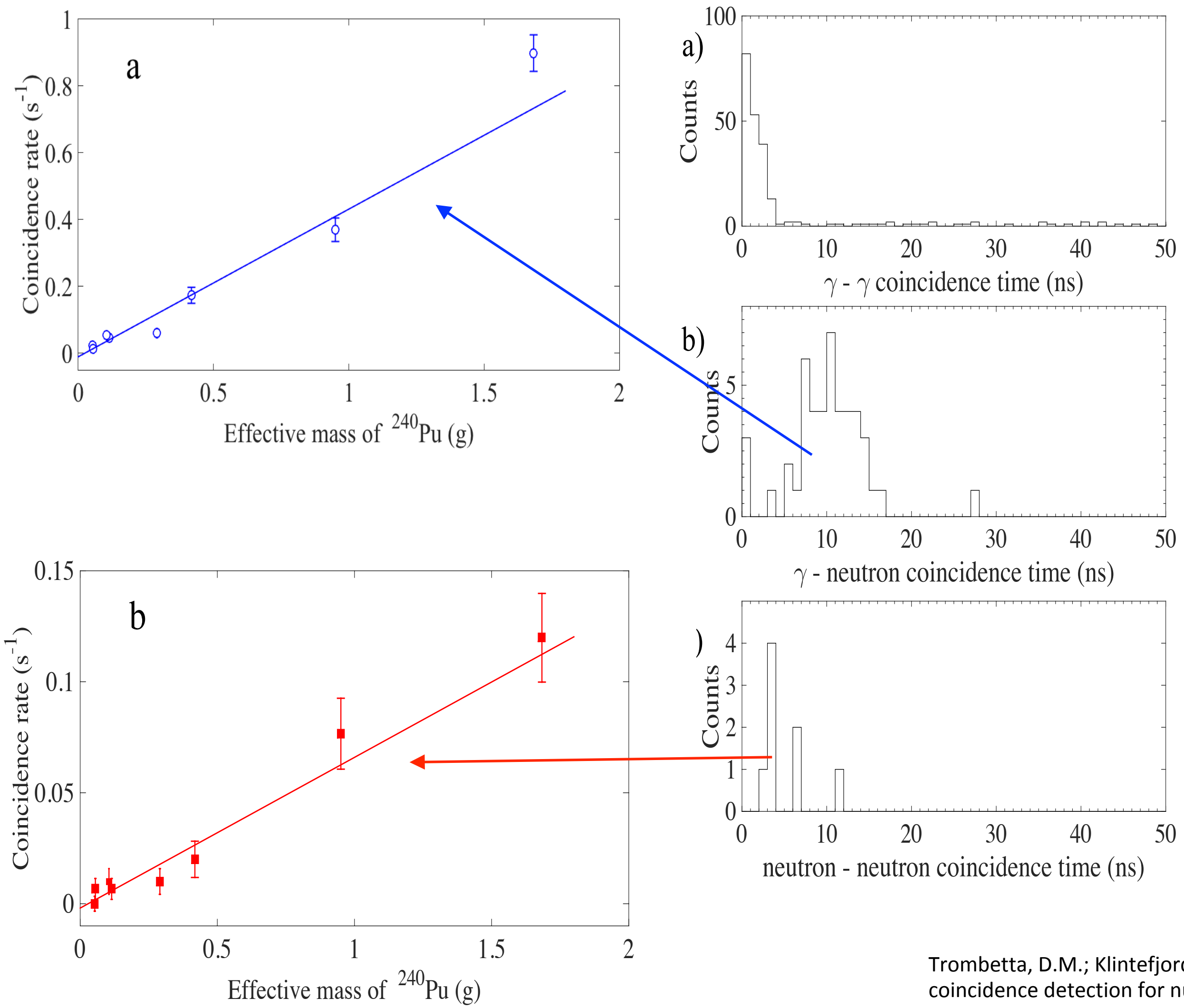
Fast neutron and gamma detection
Organic liquid scintillator detector
Monte Carlo simulations
Non-destructive analysis (NDA)
Nuclear security
Nuclear safeguards

ABSTRACT

The use of passive and active interrogation techniques to evaluate materials concerning their content of special nuclear materials (SNM) is fundamental in fields such as nuclear safeguards and security. Detection of fast neutrons and γ rays, which are a characteristic signature of SNM, has several potential advantages compared with the commonly used systems based on thermal and epithermal neutron counters, the most important being the much shorter required coincidence times and the correspondingly reduced rate of background events due to accidental coincidences. Organic scintillators are well suited for this purpose due to their fast timing properties and composition being based on carbon and hydrogen with large elastic scattering cross-sections for fast neutrons. Organic scintillators also have suitable detection efficiency for γ rays and exhibit pulse shape properties which are favorable for distinguishing between neutrons and γ rays. This paper presents experimental results and Monte Carlo simulations for a neutron–neutron and γ-neutron coincidence detection setup for identification and characterization of SNM based on such detectors. The measurements were carried out on different samples of PuO_2 material with varying content of ^{240}Pu at the Joint Research Center (JRC) of the European Commission, Ispra, Italy. The results demonstrate significant advantages of fast neutron-γ coincidence detection over fast neutron–neutron coincidence counting for certain applications, e.g. for nuclear security systems, even in the presence of moderate amounts of shielding.

Fast time correlations for enhanced detection of SNM



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Monte Carlo

- **Validation**
- **Design**

MCNP(v6.2) INPUT

*Geometry setup
* sample
* detectors

Physics

*Source:
SF and SP
FMULT CARD – CGMF and FREYA codes

Tally
* PTRAC CARD

→

MCNP OUTPUT

- Type of Interaction
- Particle
- Energy
- Time
- Momentum

→

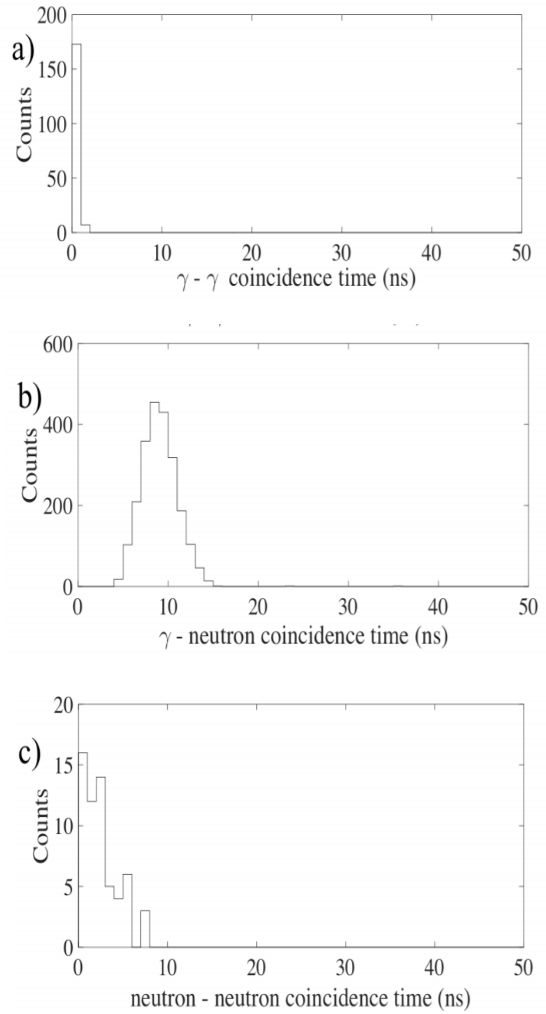
MATLAB

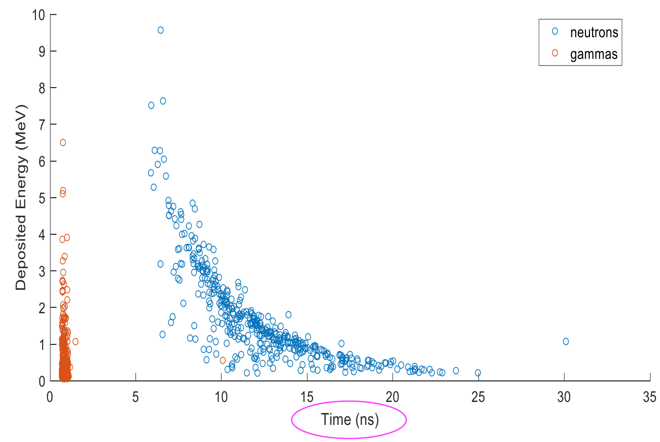
- Coincidence couting
- Energy Deposition
- Time of flight

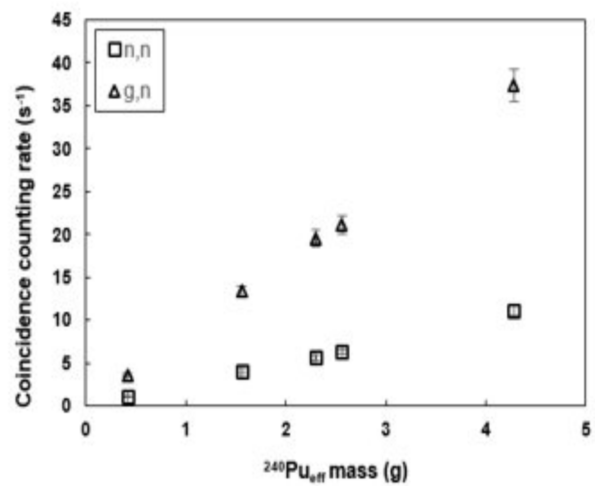
Note: Prior to the inclusion of the LLNL Fission Library, all photons produced from all neutron reaction channels were sampled prior to the selection of the neutron reaction, meaning that gamma rays could not be correlated with specific neutron reactions actually taking place in the simulation

Fast neutron-gamma correlations for sensitive detection of SNM

- Coincidence time spectra

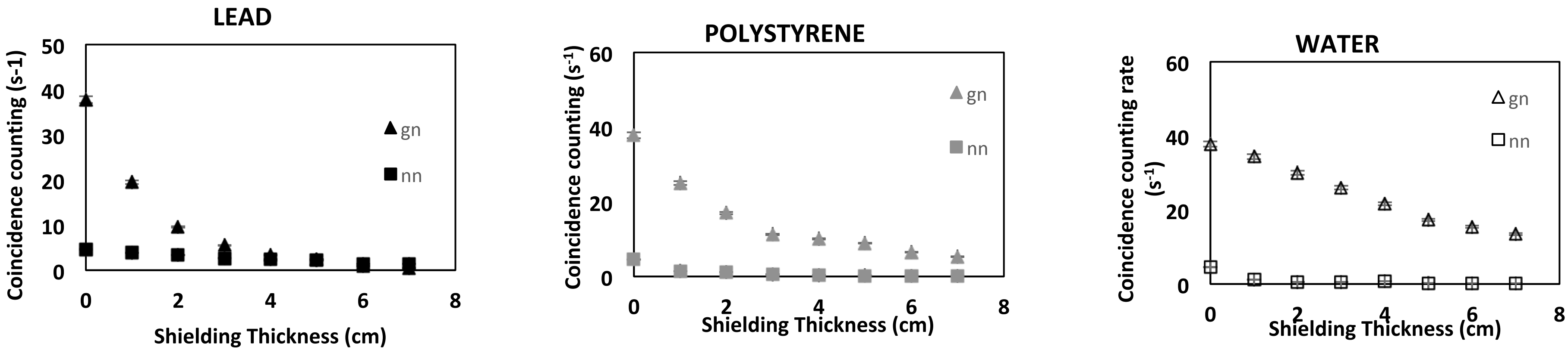


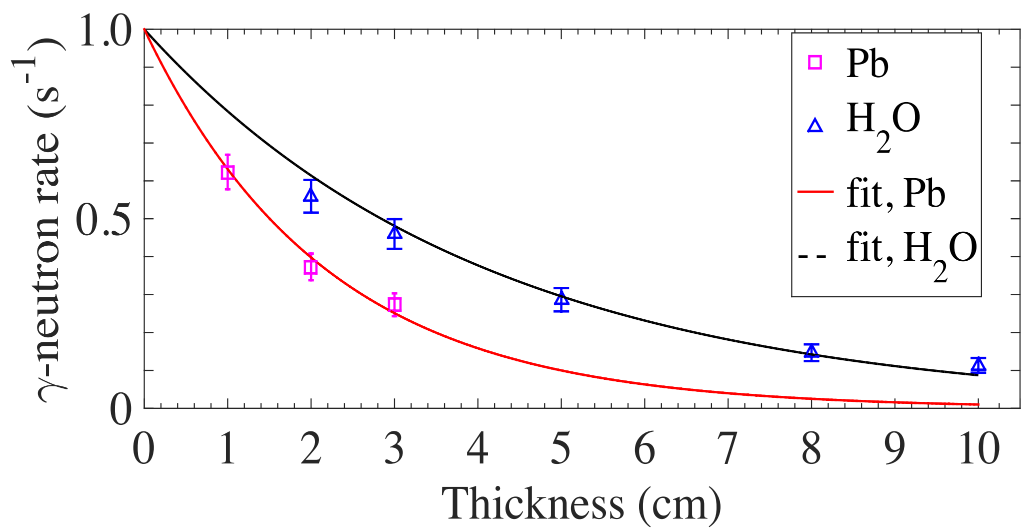




coincidence counting rates as function of $^{240}Pu_{eff}$ mass

- γ-neutron and neutron-neutron coincidence rates for (a) lead, (b) polystyrene and (c) water shielding as a function of shielding thickness.





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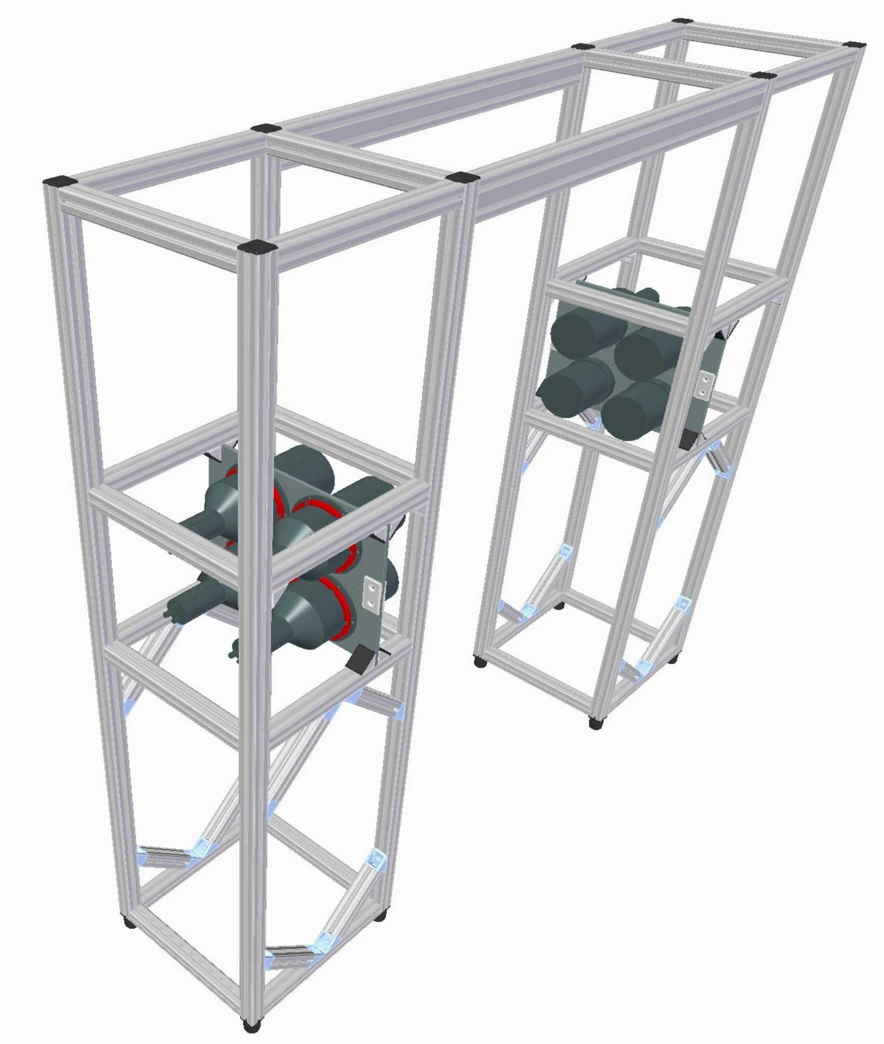
RPM Prototype development



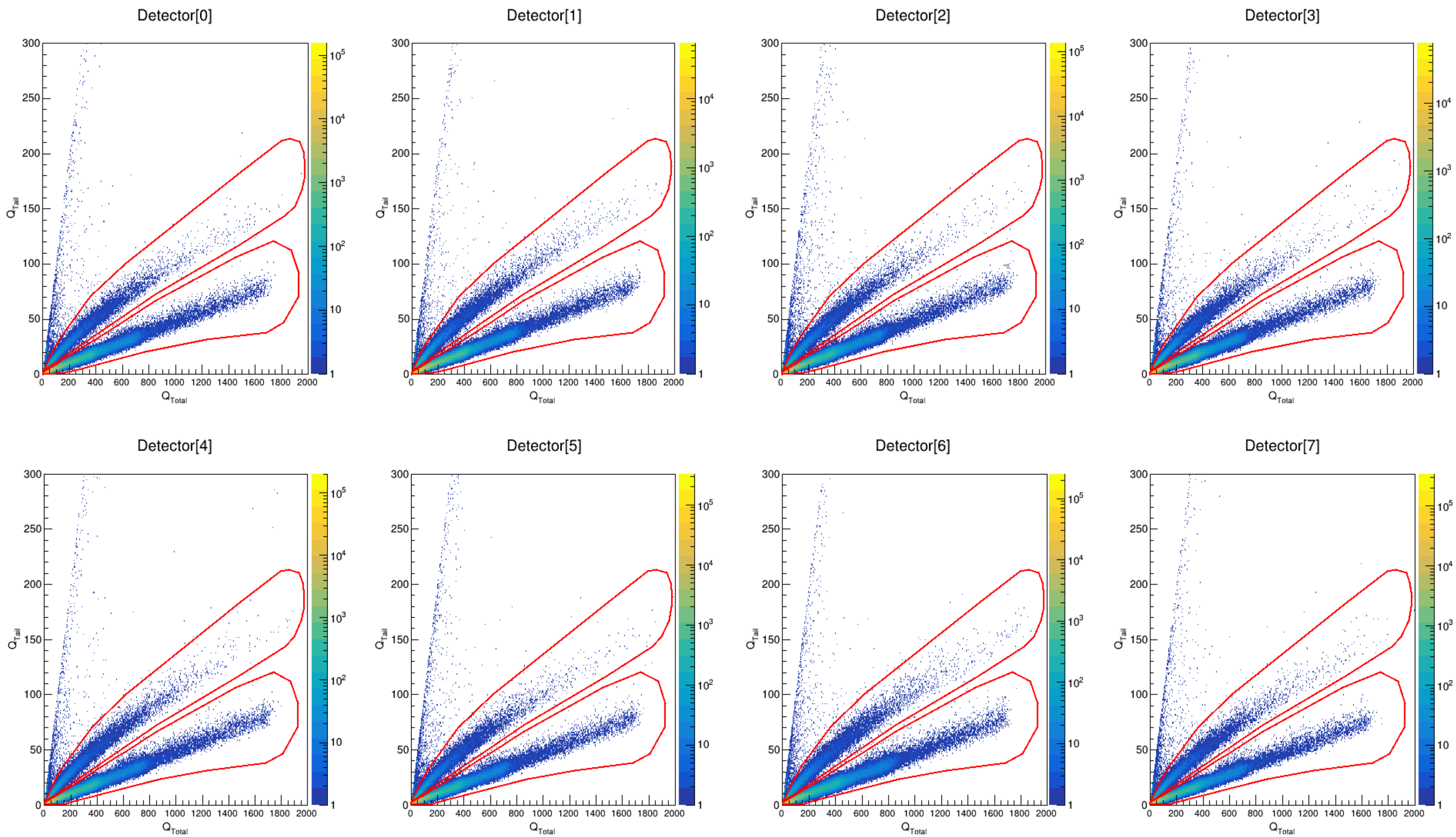
RPM Prototype development

8x EJ-309 127 cm2 x 13 cm liquid scintillation detectors

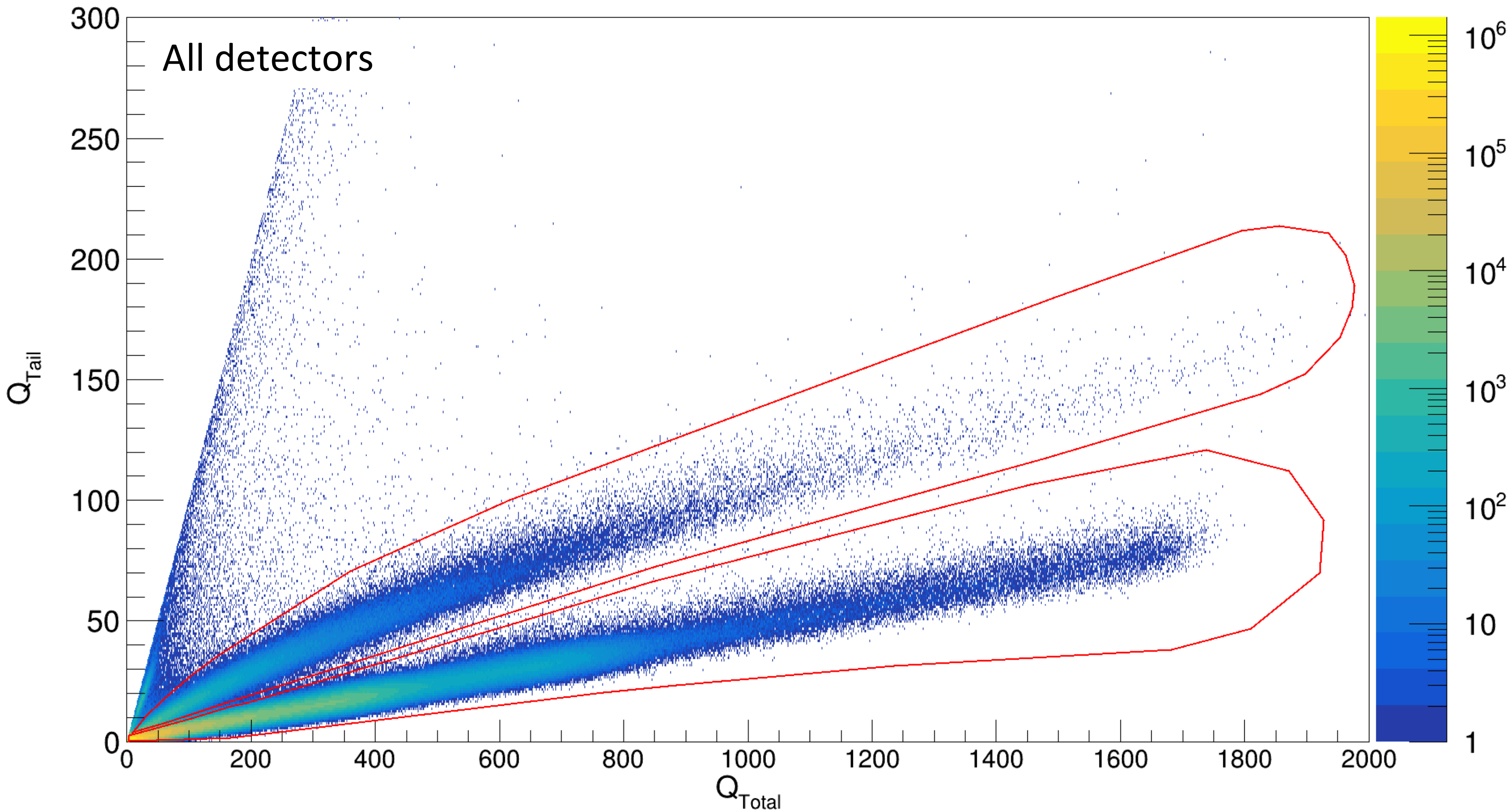
DACQ based on high-speed digitizers

- 8x 14 bit, 500 MHz
- Online n/γ PSD

RPM prototype data - PSD

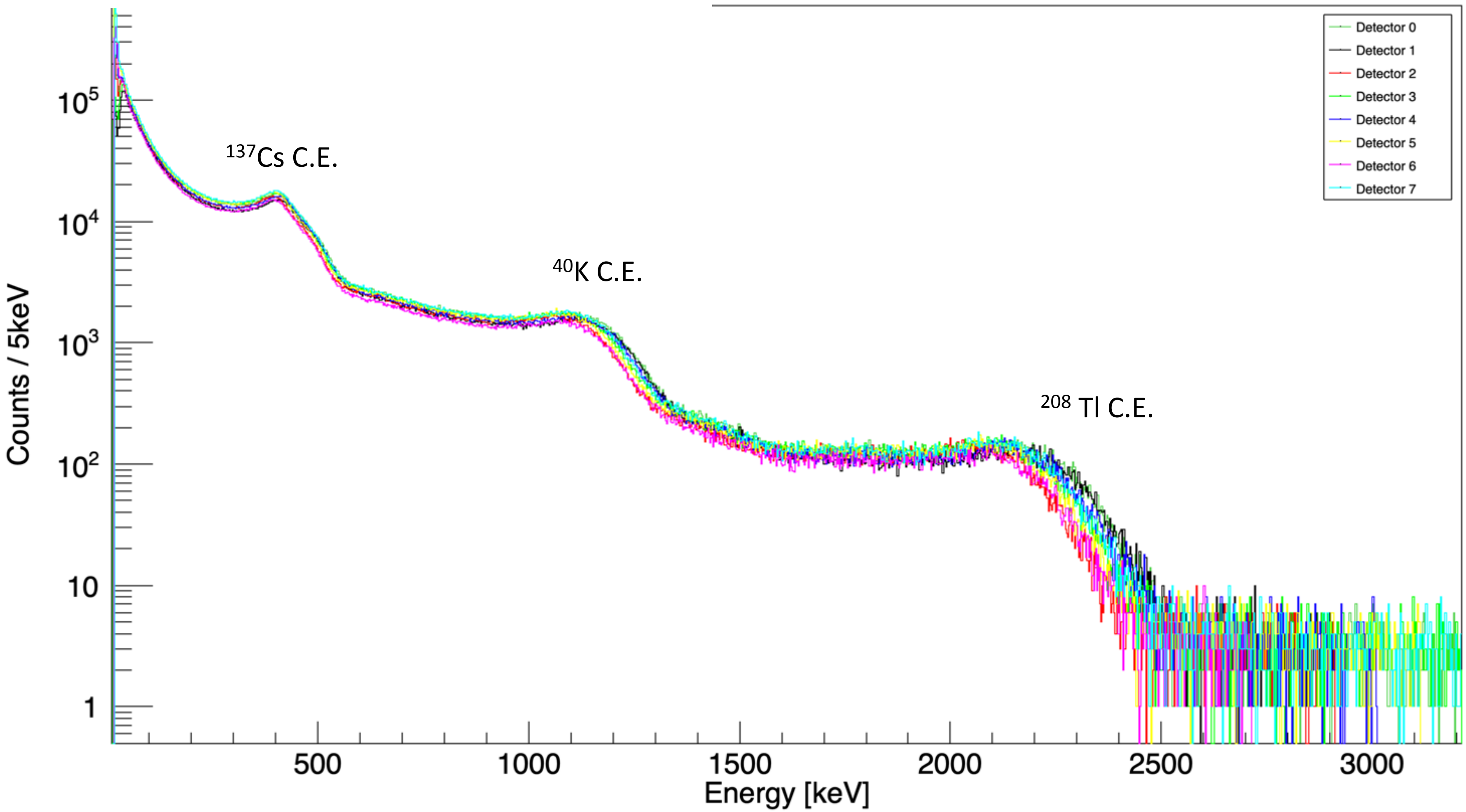


RPM prototype data - PSD



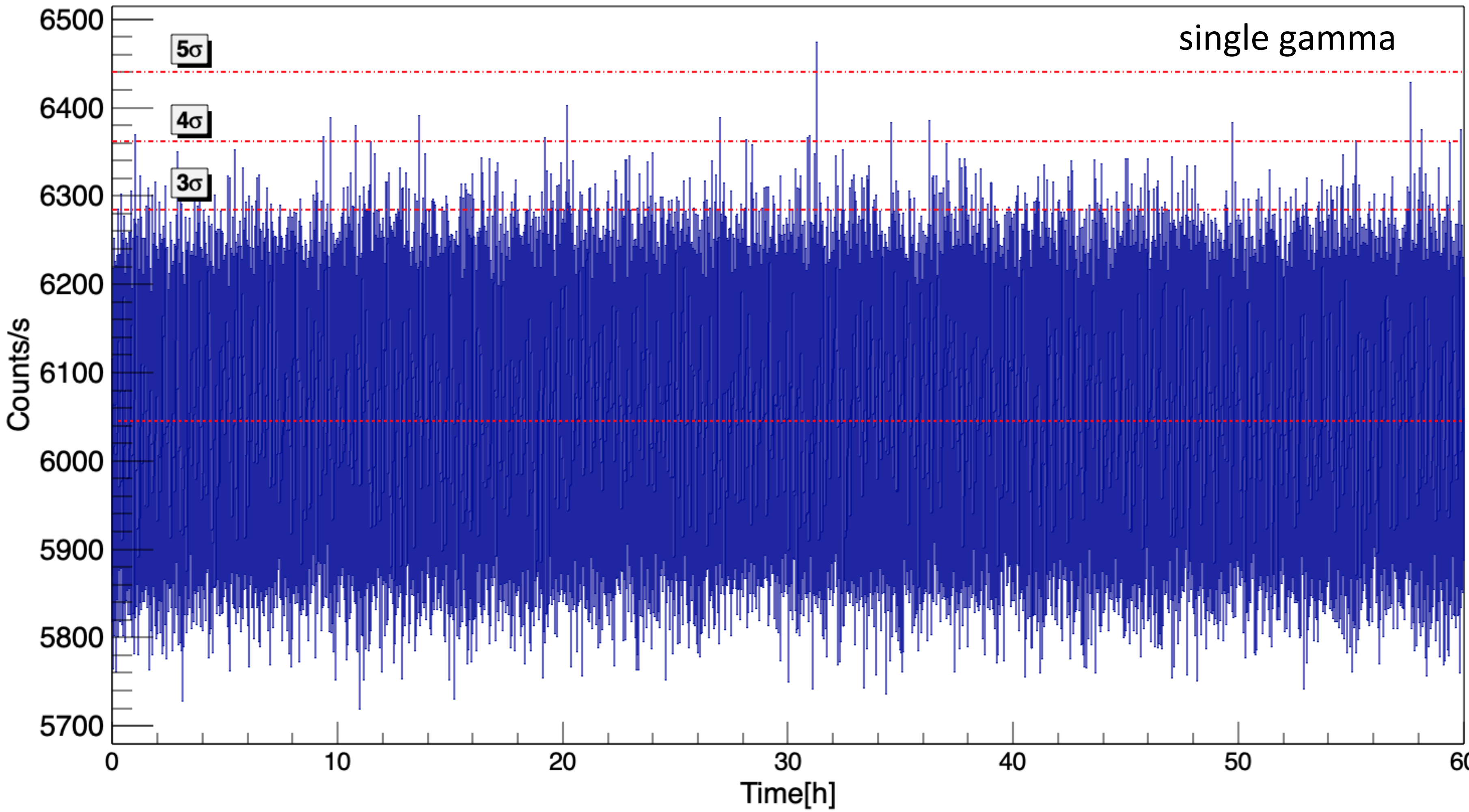
RPM prototype - ^{137}Cs calibration

All detectors – uniform signal properties



RPM prototype
60 h background test (KTH detector lab)

216000 1s measurements



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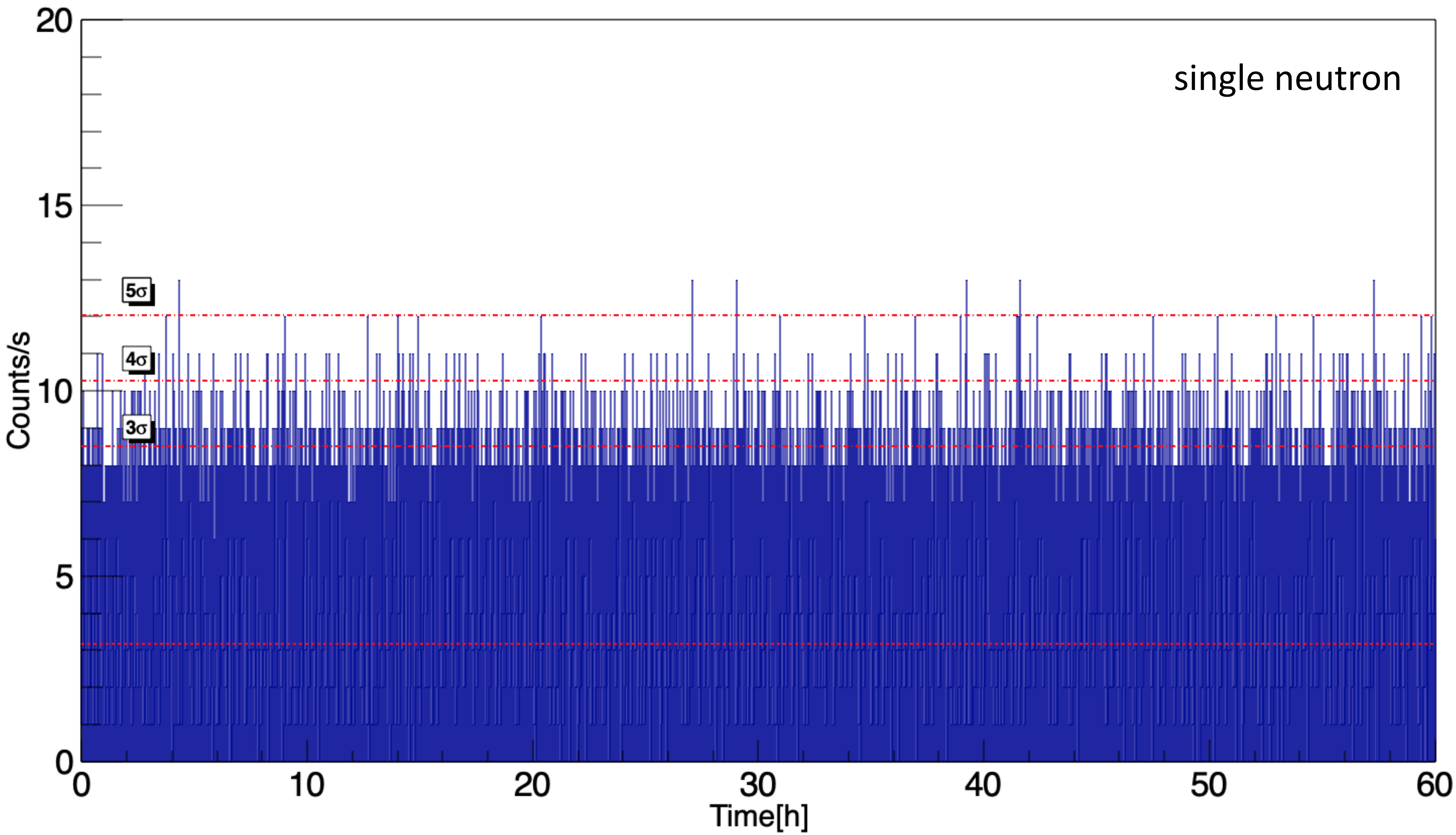

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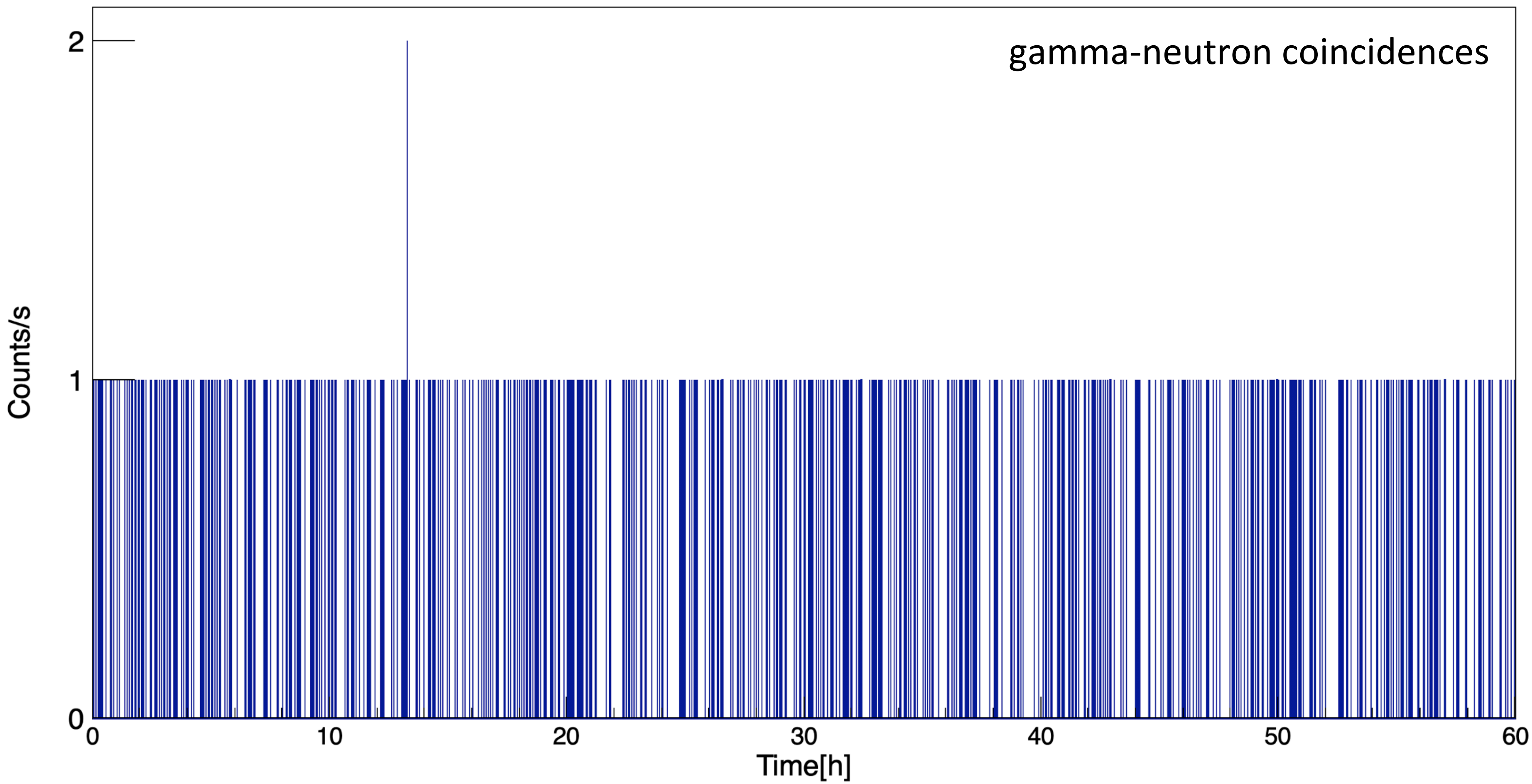


RPM prototype simulations (MCNP6.2)
ANSI N42.35-2016 standard ^{252}Cf neutron source and beyond (50%)
Single-passage, 1s interrogation, 1.2m/s, 1m above floor

ALARM TESTS FOR ANSI STANDARD AND BEYOND
"BARE" ^{252}Cf (1 cm STEEL, 0.5 cm LEAD SHIELDING).

SOURCE NEUTRON EMISSION RATE	SINGLE NEUTRONS N / p_N	GAMMA-NEUTRON COINCIDENCES N / p_N	NEUTRON-NEUTRON COINCIDENCES N / p_N
20 000 n/s	150 / $<10^{-12}$	184 / 0.0015	46 / 0.013
10 000 n/s	75 / $<10^{-12}$	92 / 0.030	23 / 0.096

ALARM TESTS FOR ANSI STANDARD AND BEYOND
MODERATED ^{252}Cf (4 cm HDPE SHIELDING).

SOURCE NEUTRON EMISSION RATE	SINGLE NEUTRONS N / p_N	GAMMA-NEUTRON COINCIDENCES N / p_N	NEUTRON-NEUTRON COINCIDENCES N / p_N
20 000 n/s	102 / $<10^{-12}$	185 / 0.0014	12 / 0.27
10 000 n/s	51 / $<10^{-12}$	93 / 0.029	6 / 0.54

RPM prototype source tests
Single-passage, 1s interrogation, 1.2m/s, 1m above floor

VINNOVA

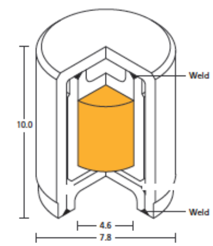
Strål säkerhets myndigheten
Swedish Radiation Safety Authority

ALARM TESTS FOR ^{252}Cf – MEASUREMENT.

SOURCE NEUTRON EMISSION RATE	SINGLE NEUTRONS N/p_N	GAMMA-NEUTRON COINCIDENCES N/p_N	NEUTRON-NEUTRON COINCIDENCES N/p_N
8 300 n/s	68 / $<10^{-12}$	123 / 0.011	20 / 0.12

42% of ANSI N42.35-2016

X.1



ALARM TESTS FOR GAMMA SOURCES: ^{137}Cs and ^{133}Ba – MEASUREMENT.

SOURCE	ACTIVITY	SIGMA MULTIPLIER (N)	PROBABILITY OF FALSE NEG. (p_N)
^{133}Ba	50 kBq	11	$<10^{-12}$
^{137}Cs	191 kBq	31	$<10^{-12}$

10% of ANSI N42.35-2016

32% of ANSI N42.35-2016

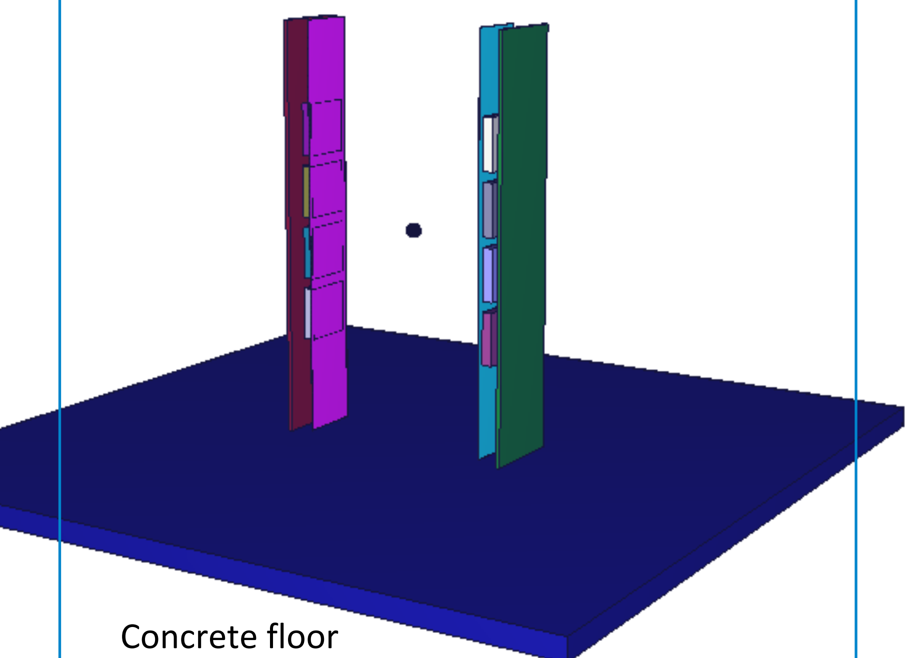
ALARM TRIGGER THRESHOLDS (>4σ above background)

	counts/s
Single neutrons	10
Single gamma rays	6340
Gamma-neutron coinc.	1
Neutron-neutron coinc.	1

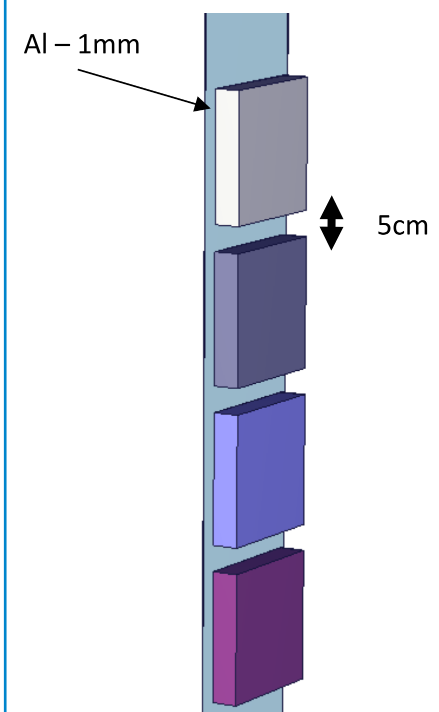
MCNP6.2 SIMULATIONS FOR NOVEL RPM DESIGNS AND DETECTOR MATERIALS

RPM DESIGN – PLASTIC SCINTILLATORS

DOUBLE SIDED RPM

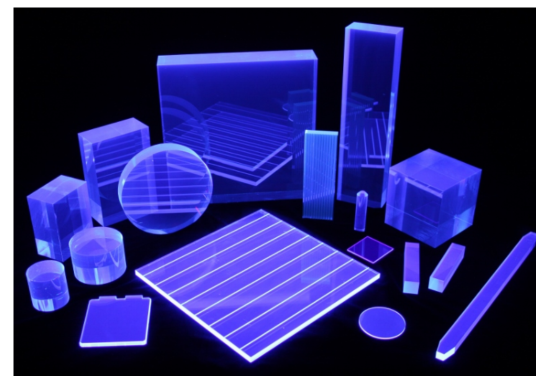


DETECTION ASSEMBLY

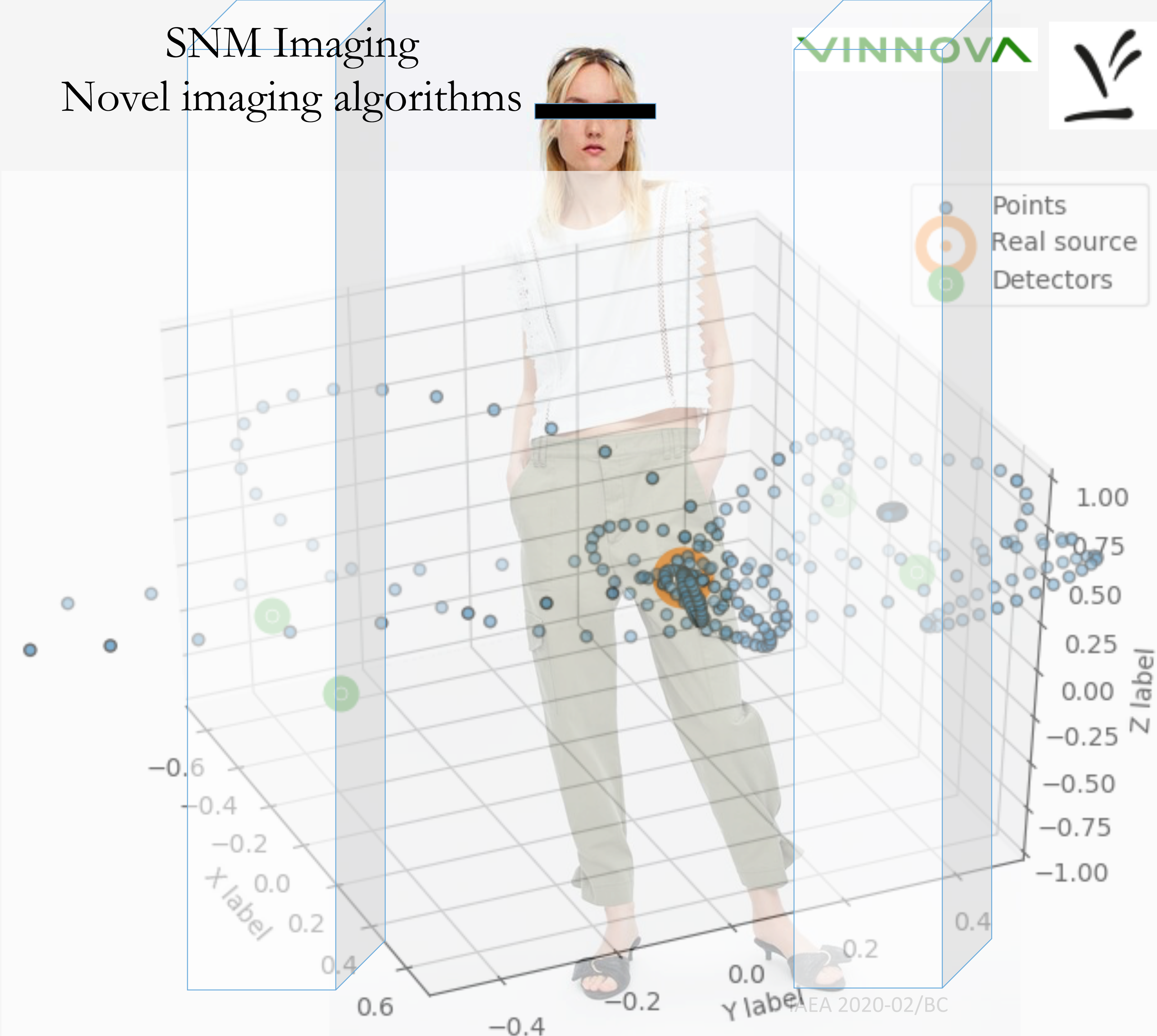


DETECTOR EJ-299

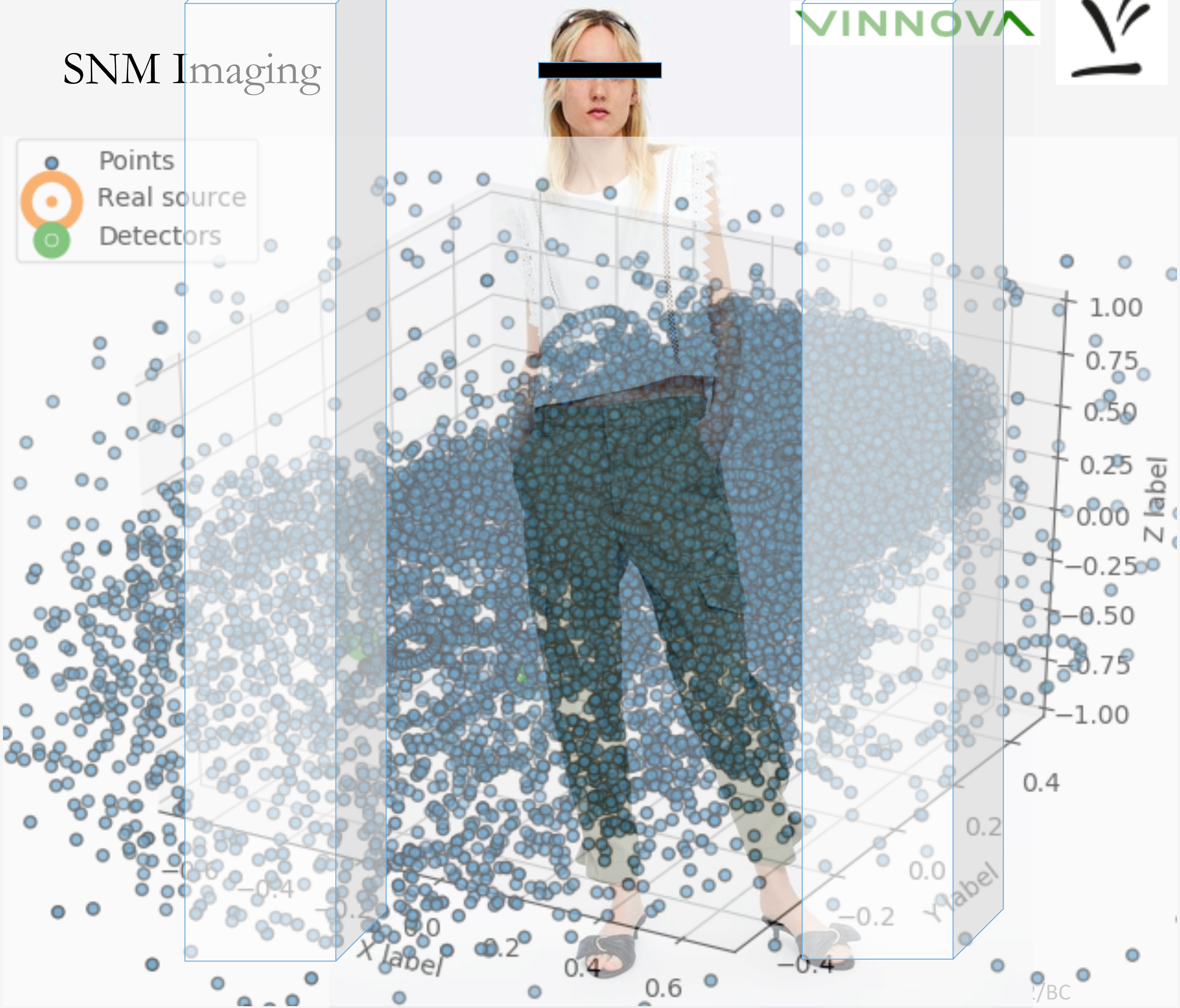
Area: 25cm x 25cm
Thickness: 25cm



SNM Imaging
Novel imaging algorithms

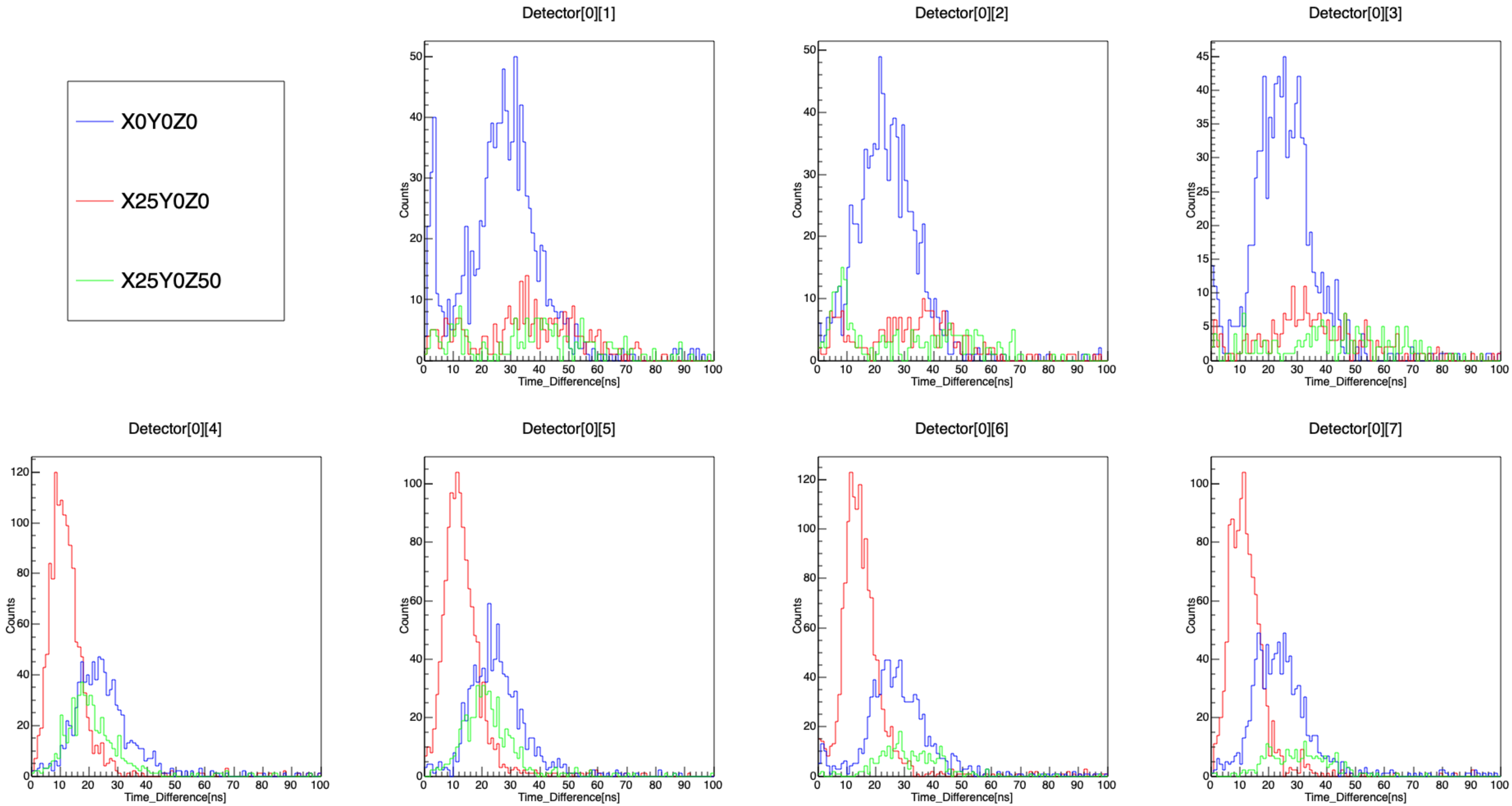


SNM Imaging

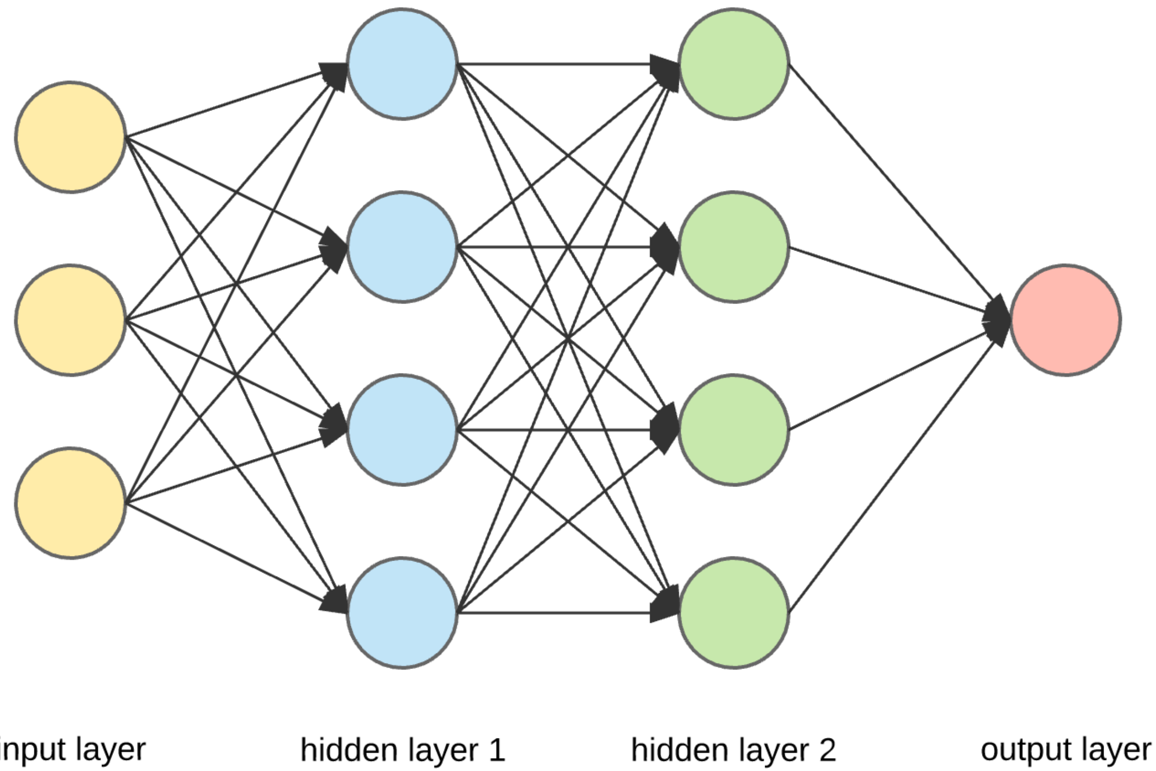


Points
Real source
Detectors
1.00
0.75
0.50
0.25
0.00
−0.25
−0.50
−0.75
−1.00
Z label
0.4
0.2
0.0
−0.2
−0.4
Y label
−0.6
−0.4
0.0
0.2
0.4
0.6
X label

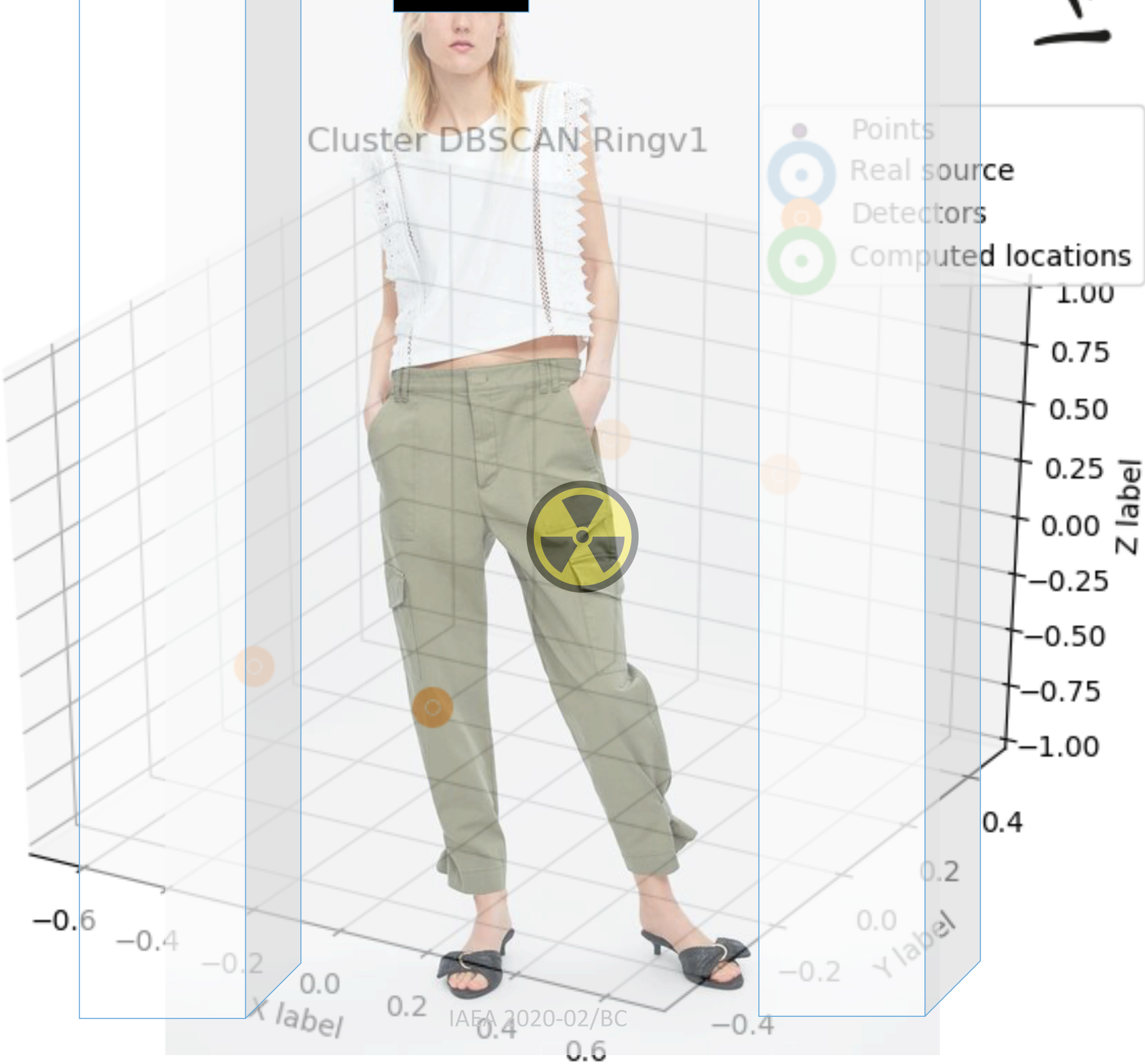
RPM prototype data – novel imaging modality



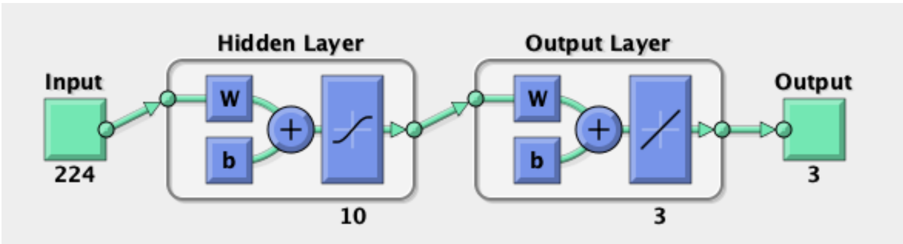
SNM Imaging and Deep Learning

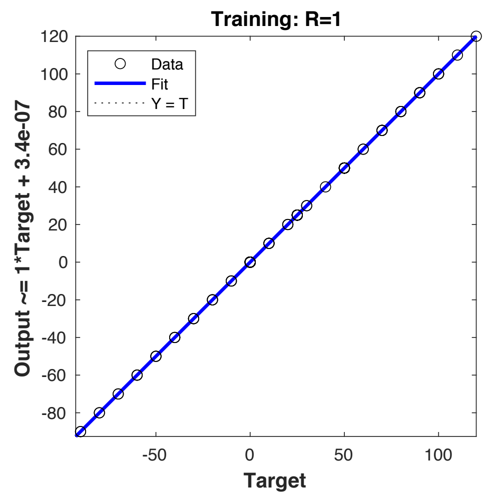


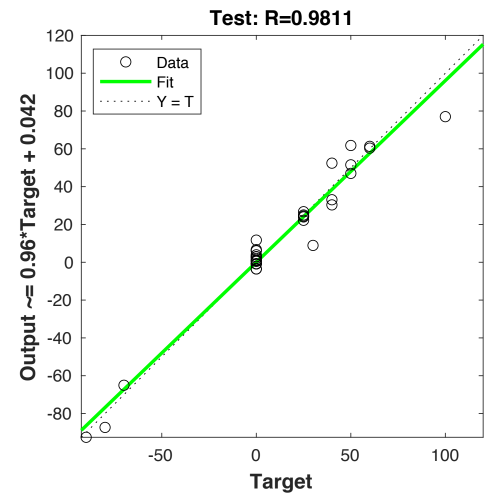
SNM Imaging

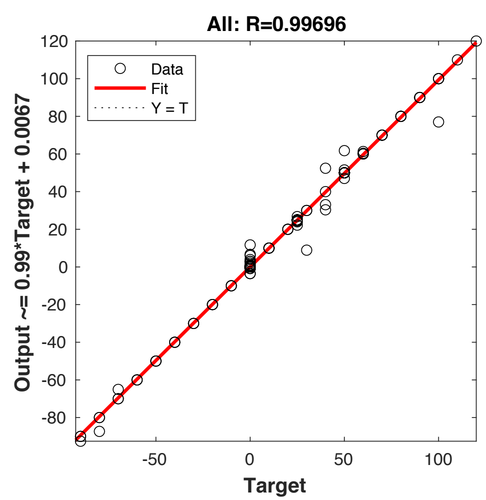

Cluster DBSCAN Ringv1
Points
Real source
Detectors
Computed locations
Z label
1.00
0.75
0.50
0.25
0.00
−0.25
−0.50
−0.75
−1.00
X label
−0.6
−0.4
−0.2
0.0
0.2
0.4
0.6
Y label
0.4
0.2
0.0
−0.2
−0.4

Preliminary results









Summary

Fast γ-neutron coincidence counting has been developed as a novel sensitive tool to detect SNM for applications in Nuclear Security (RPMs), Nuclear Safeguards (NDA in passive and active interrogation scenarios), Environmental Surveying and Emergency response.

- The method is inspired by techniques used in state-of-the-art nuclear physics experiments using arrays of organic scintillation detectors, high-speed sampling ADCs ("digitizers"), fast timing, and pulse processing algorithms for discriminating between neutrons and γ-rays.
- Complements normal singles gamma and neutron alarming in different RPM applications
- A limited scale (13 l organic scintillator, 8 modules) pedestrian/package/luggage RPM has been developed at KTH as proof of concept
- Limited spectral capabilities for radionuclide identification (mainly Compton reconstruction)
- "Intelligent" alarm trigger for suppression of NORM/medical nuisance alarms
- Enhanced SNM imaging capabilities (beyond intensity mapping) based on fast neutron-gamma correlations
- Validated (ANSI N42.35-2016) and for quantifying small amounts (ranging from 0.5g to 1.5g) of ^{240}Pu.
- Int. PCT Patent application No PCT/SE2019/050609

<u>Other ongoing and planned technology developments</u>

- Other RPM applications (vehicle, etc)
- Development of imaging algorithms
- "In-situ" spent fuel verification and imaging before final repository
- Environmental and emergency response