Characterization of ^{226}Ra implanted sources for background mitigation and high-sensitivity α spectroscopy

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Low **R**adioactivity **T**echniques
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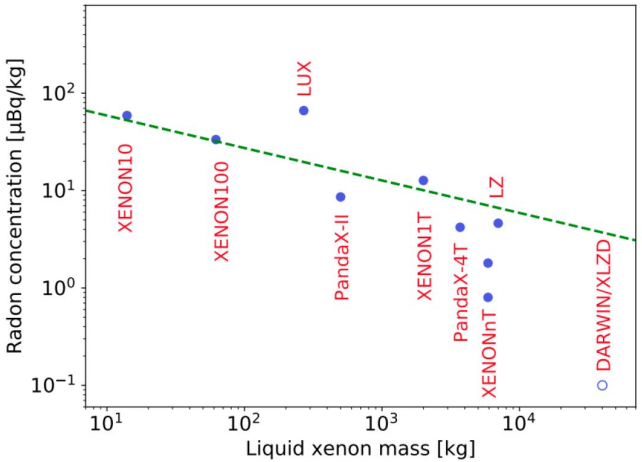
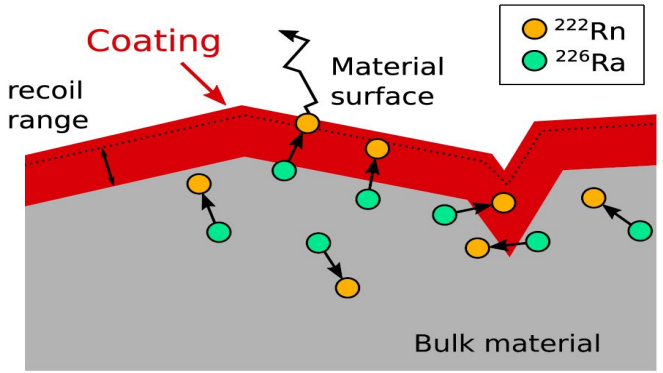
^{222}Rn Background in XLZD: Motivation for Surface Coating

- ^{222}Rn enters the detector via emanation from construction materials, daughters β-decays (^{214}Pb, ^{214}Bi) continuous spectra → overlap with WIMP ROI
- Rn emanation ∝ surface area → concentration scales as ~ **$m^{-1/3}$** → simply increasing detector mass **does not solve the problem**



- **Coating:** thin layer applied to material surfaces to **suppress ^{222}Rn emanation**
- Must contain the Rn recoil range (~tens of nm) and block diffusion
- Promising method: **electrochemical copper plating (ECP)** [1], [2]
- Previous result: **Rn reduction >= 1000**



[1] F. Jörg, G. Eurin, H. Simgen, Application of surface coating for radon mitigation in rare-event [2] See also M. Noia, S. Armbruster, this conference

^{226}Ra Implanted samples at ISOLDE

Production & Source

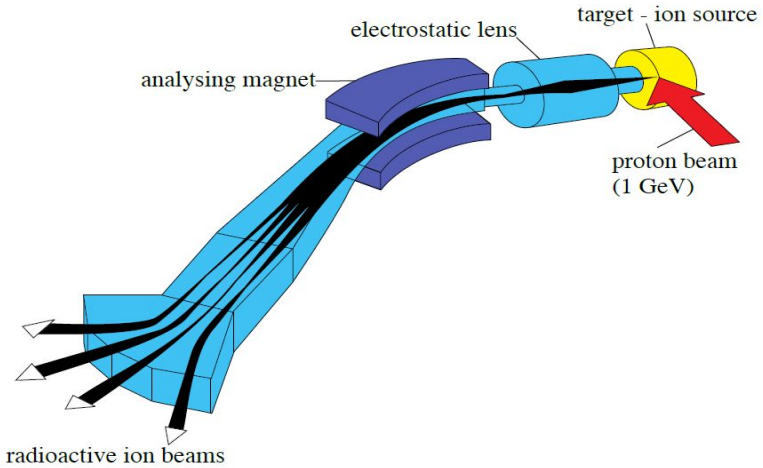
- ^{226}Ra produced via proton bombardment of Thorium Carbide target (~10^{18} protons)
- Long half-life → **off-line implantation** (no new irradiation)

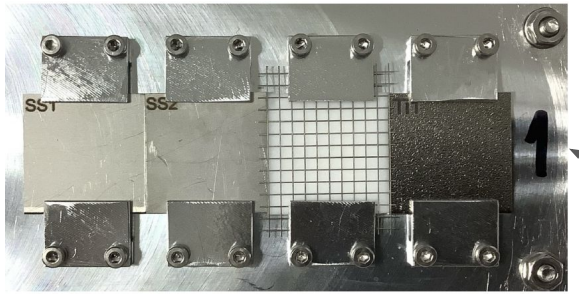
Beam Preparation

- ^{226}Ra released by **thermal diffusion**
- **Surface ionized** and accelerated to **40-50 kV**
- Purified with **magnet separator**

Implantation Setup

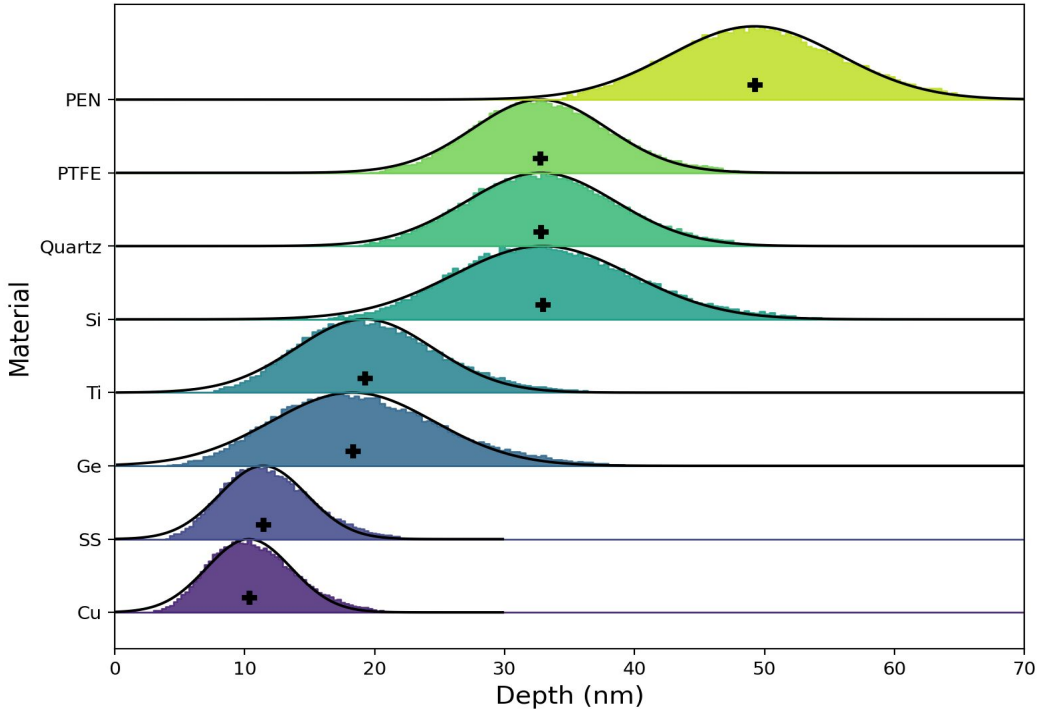
- Samples (5 mm, 2×2 cm²)
- 19 samples, 9 different materials implanted (SS, Cu, Ti, Ge, Si, Quartz, PTFE, PEN, Acrylic)
- Beam scanned over **1×1 cm² area**





Implantation profile of ^{226}Ra at 40 keV with TRIDYN

- Ion irradiation simulation on solids in **dynamic mode**
- Collisional processes via BCA (Binary Collision Approximation): implantation, scattering, sputtering, point defect damage
- **μ➡ Projected Range R**: average depth at which implanted ions come to rest, depends on species, energy, density
- **σ ➡straggling**

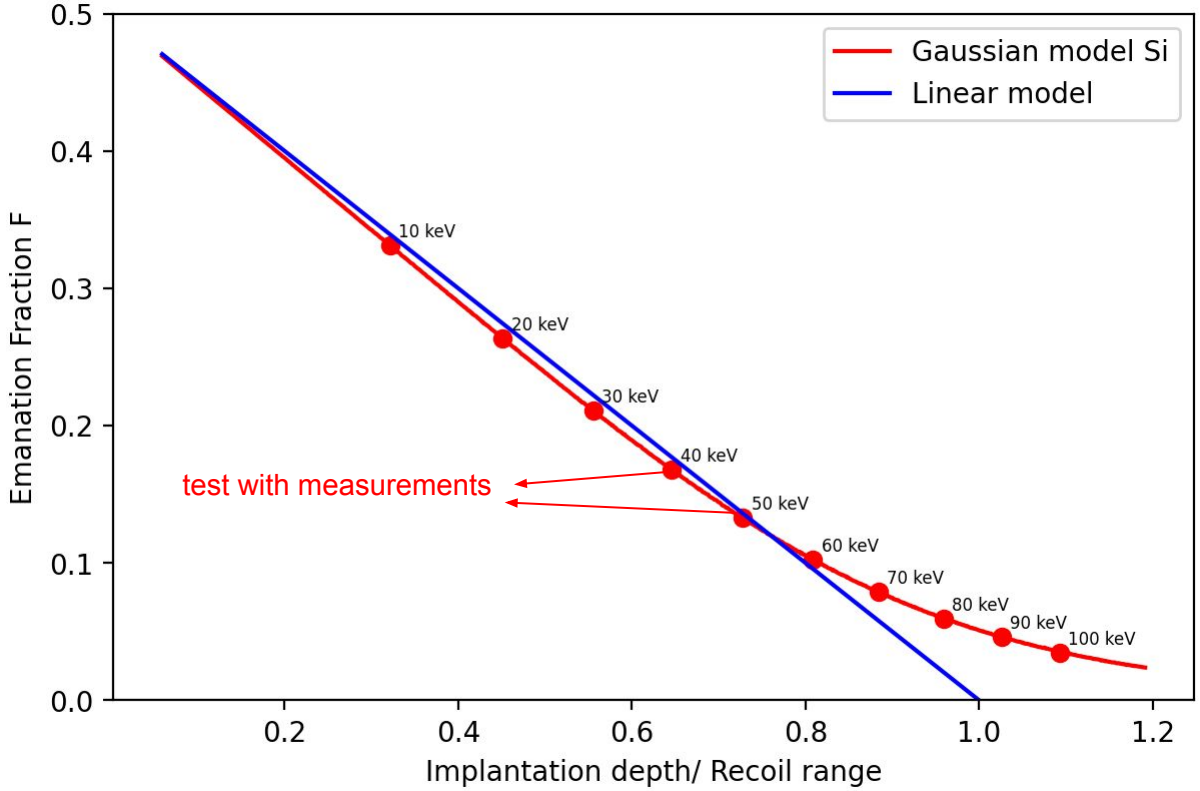


Recoil driven emanation fraction of ^{222}Rn

Emanation by recoil:

- α decay: $^{226}Ra \rightarrow ^{222}Rn + \alpha$
- TRIDYN provides the distribution of ^{226}Ra
- TRIDYN also gives the range of ^{222}Rn: 86 keV recoil energy → ~ 14nm SS, 50 nm Si
- Through geometrical considerations (linear model), we can calculate the emanation fraction

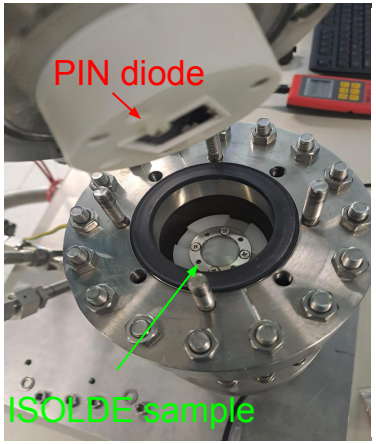
$$F = \frac{1}{2}\left(1 - \frac{z_i}{R}\right), \quad \text{for} \quad z_i \leq R$$

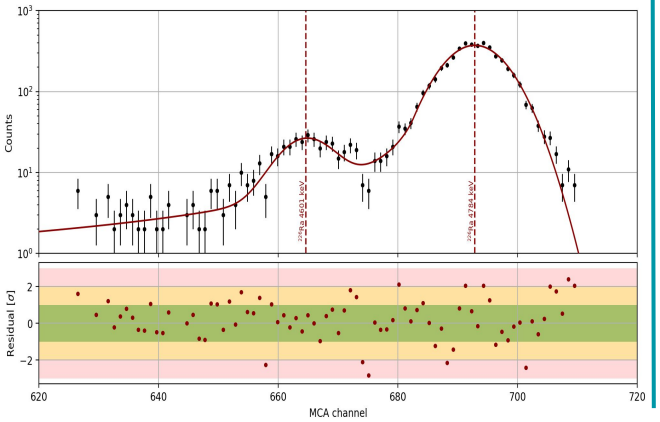


^{226}Ra activity measurements: alpha and gamma spectrometry

Alpha spectroscopy

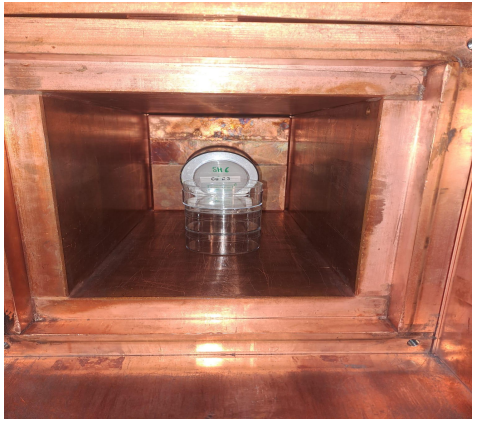
- Windowless Si PIN diode (Hamamatsu S3204-09), sample-to-diode distance adjustable → tunable geometric efficiency
- Vacuum vessel (~10^{-3} mbar) to avoid α energy loss in air
- Peaks fit with Crystal-Ball functions
- Energy resolution ≈ 60 keV FWHM at 5 MeV





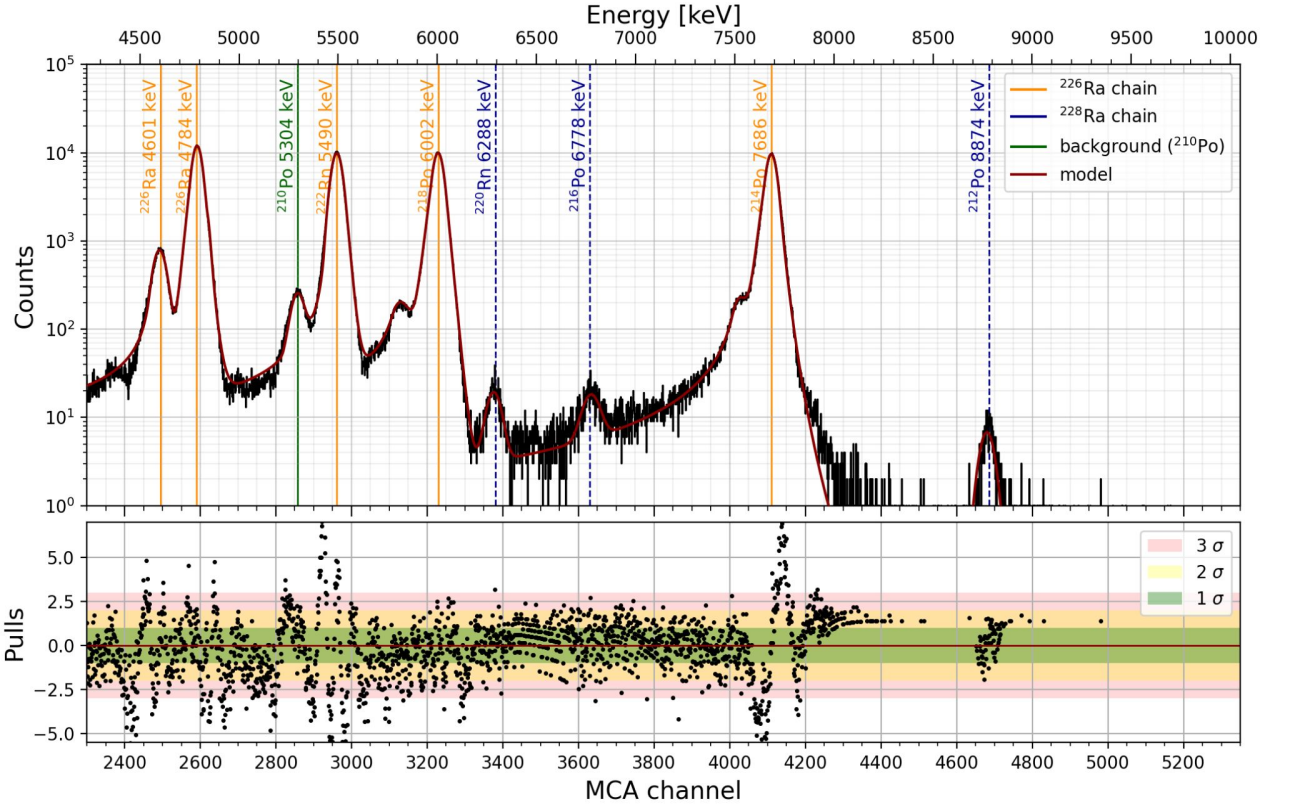
Gamma spectroscopy

- Two HPGe detectors at MPIK's Low-Level Lab: Bruno (0.64 kg crystal) and Corrado (0.94 kg, better resolution), both ~15 m.w.e. shielded, N_2-flushed
- Lower branching ratio (3.6%) + full-energy-peak efficiency correction → larger uncertainties than alpha

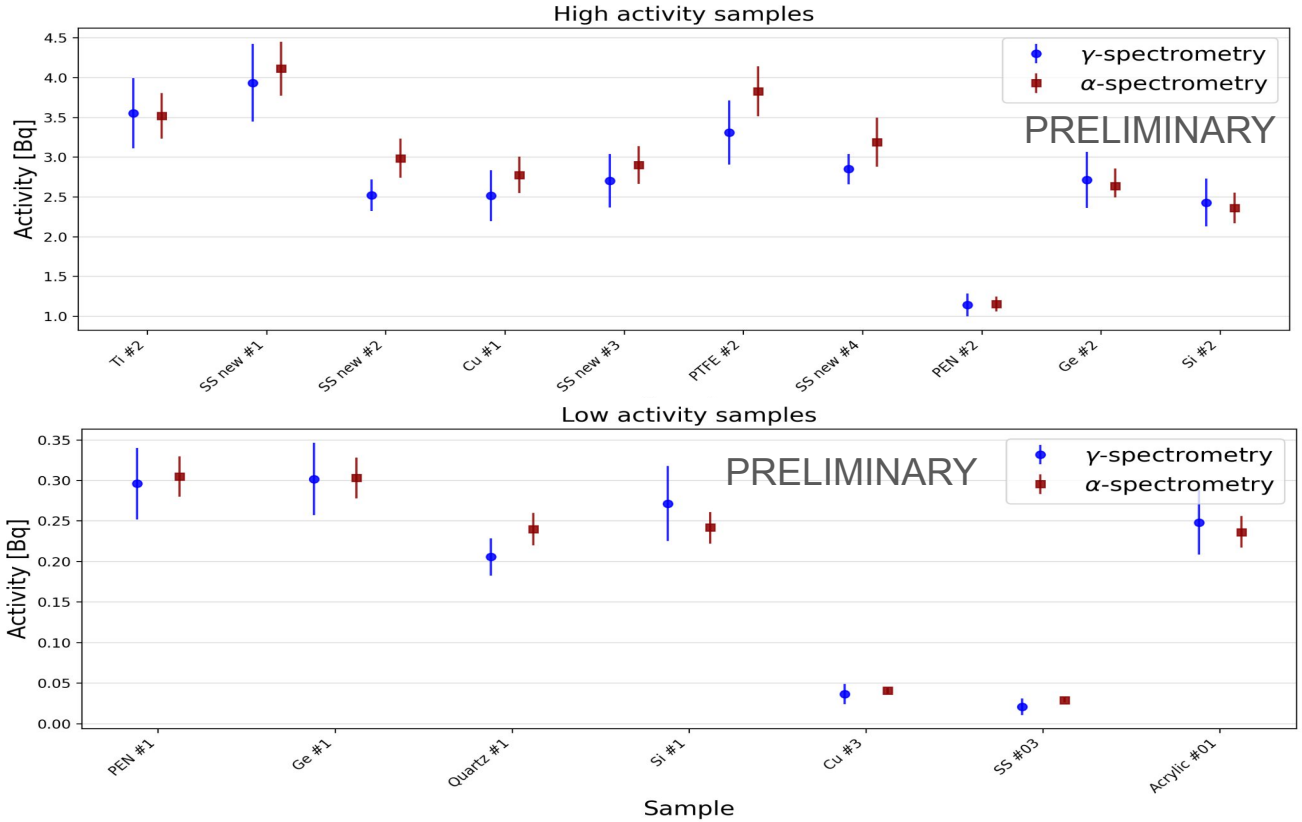


^{226}Ra activity measurements: alpha spectrometry

- **Contamination** of ^{228}Ra coming from ISOLDE implantation due to imperfect mass separation: $A_{228Ra}/A_{226Ra} = 4.54 \times 10^{-3}$
- Main **background source: ^{222}Rn and daugthers emanated** from samples can implant on detector walls → mitigated by focusing analysis on the ^{226}Ra energy range



^{226}Ra activity measurements: comparison between α and γ

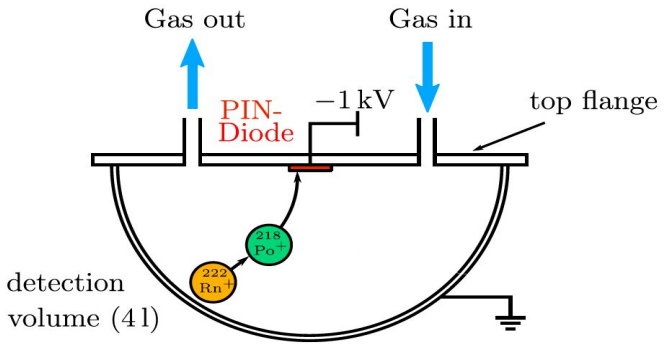


^{222}Rn emanation measurements

1. Place sample in leak tight container
2. Fill container with radon free carrier gas (e.g. N2, He, ...)
3. Wait for the radon to accumulate → $A(t) = A_{eq} \cdot \left(1 - e^{-\lambda_{222Rn} \cdot t}\right)$

Electrostatic Radon Monitor

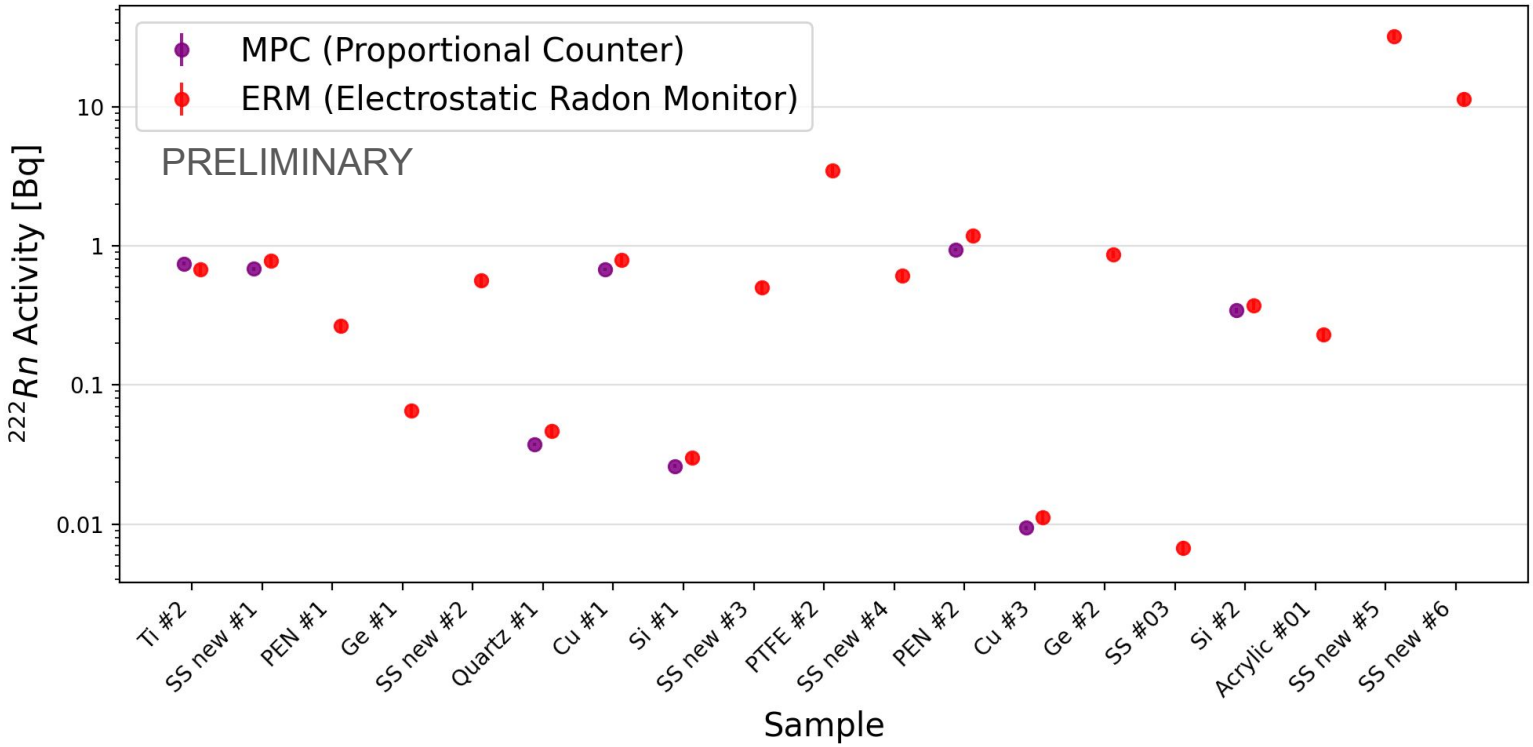
- Emanated ^{222}Rn transferred into detector
- Daughter ions (^{218}Po, ^{214}Po) drift onto a biased Si PIN diode → counted by α-spectroscopy
- Calibrated with a reference ^{226}Ra source →



Proportional Counters

- Heritage: developed at MPIK for GALLEX; ~1 cm³ active volume, hand-crafted fused silica
- Radon transferred into counter, filled with P10 (90% Ar / 10% CH_4); α-decays counted directly
- Sensitivity down to 20 μBq (20% uncertainty) → ideal for the lowest-activity samples

^{222}Rn emanation measurements: results ERM & MPC



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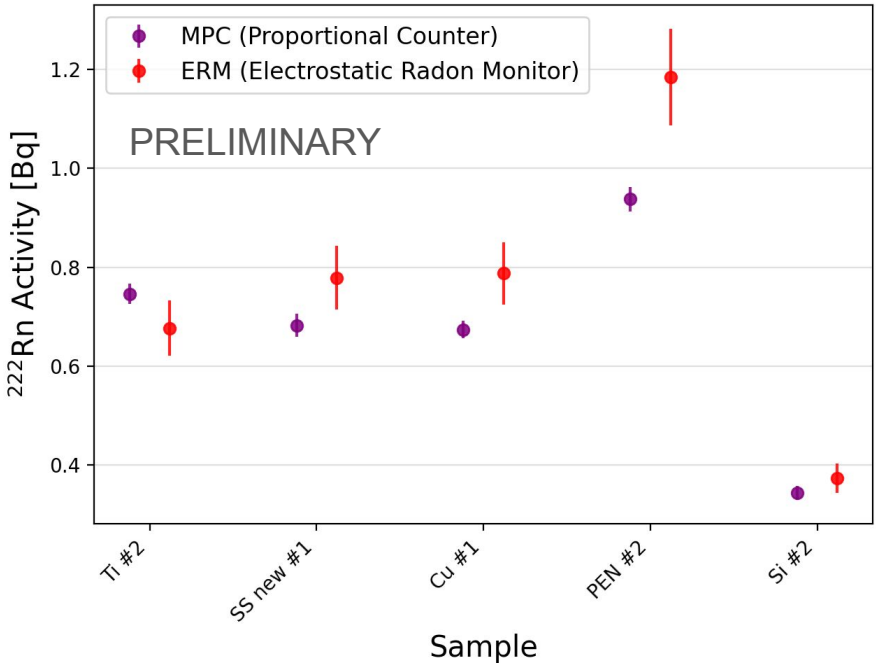
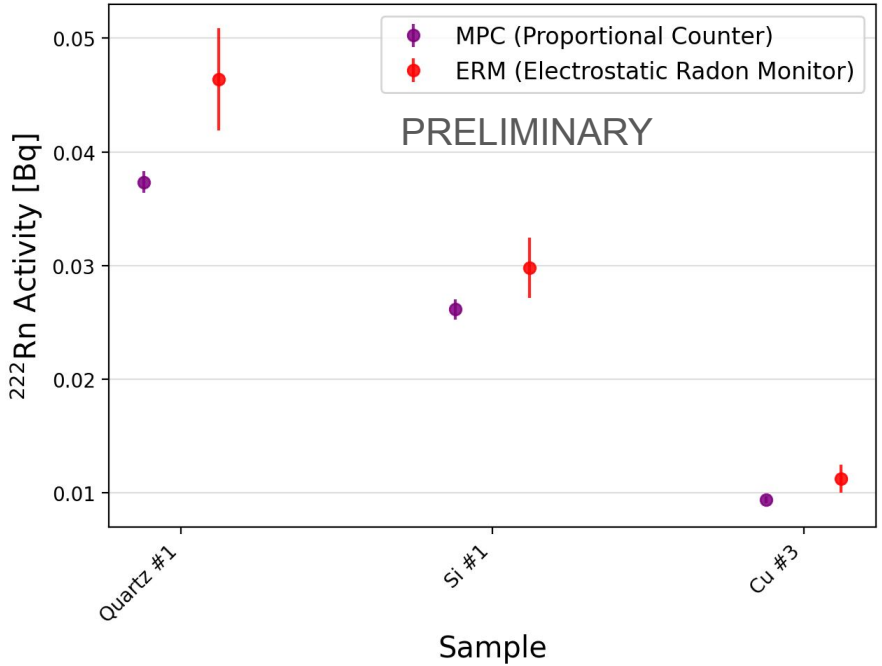
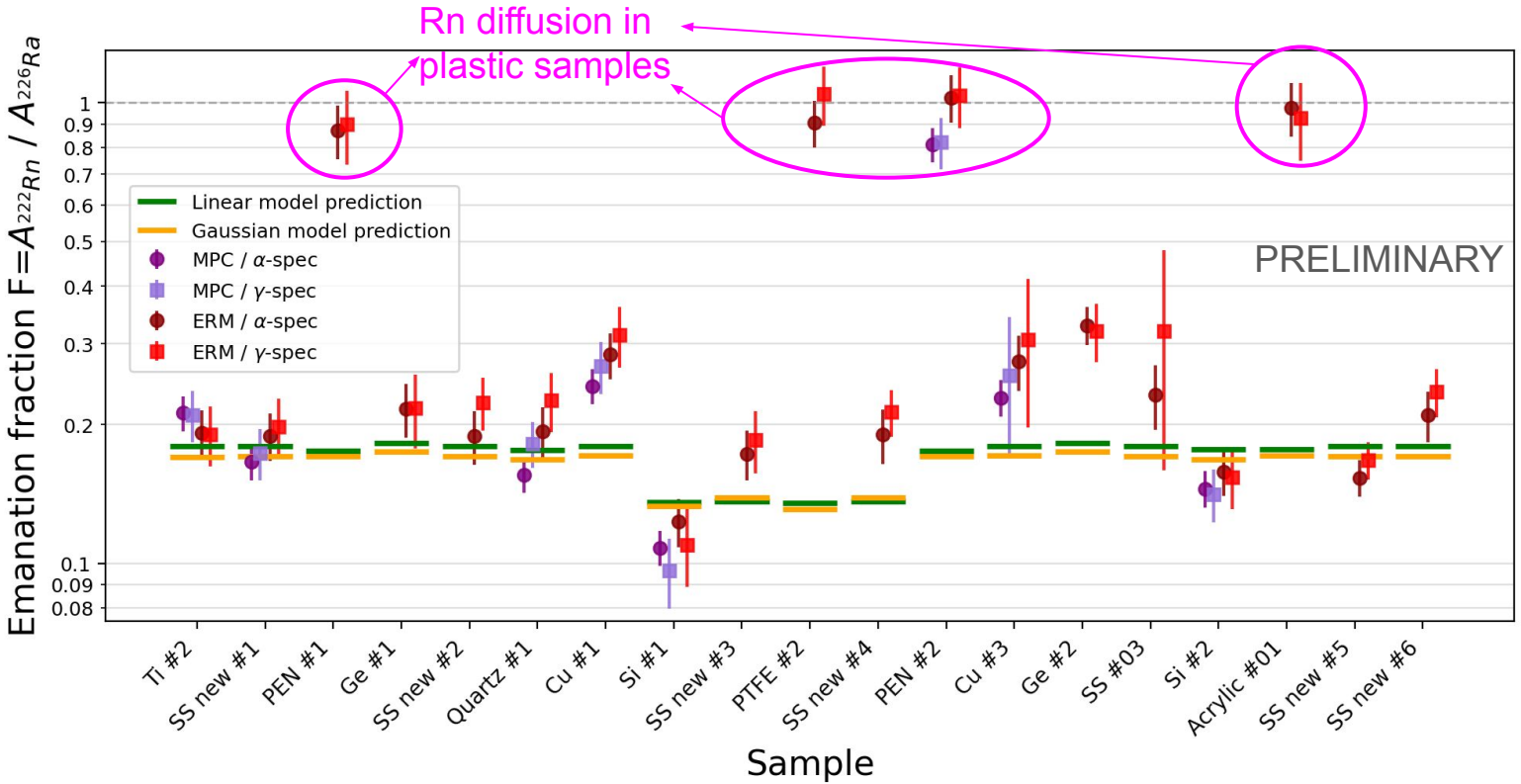




Slight tension between MPC and ERM measurements → possible systematic bias

^{222}Rn emanation measurements: comparison with simulations



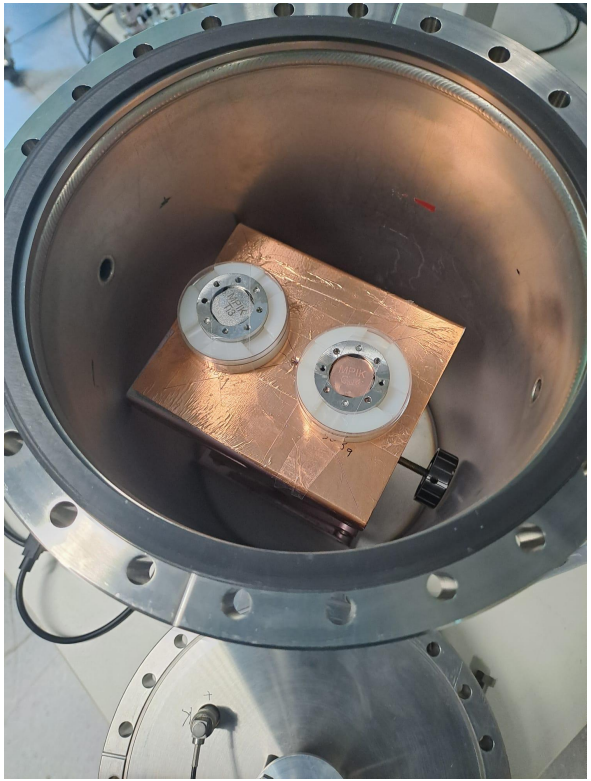
^{222}Rn diffusion coefficient measurement in crystalline solids

Why measuring Rn diffusion in detector materials?

- Diffusion coefficients in metals are **unknown**, no systematic measurements exist
- Knowledge of D allows to **predict and mitigate** Rn-induced backgrounds

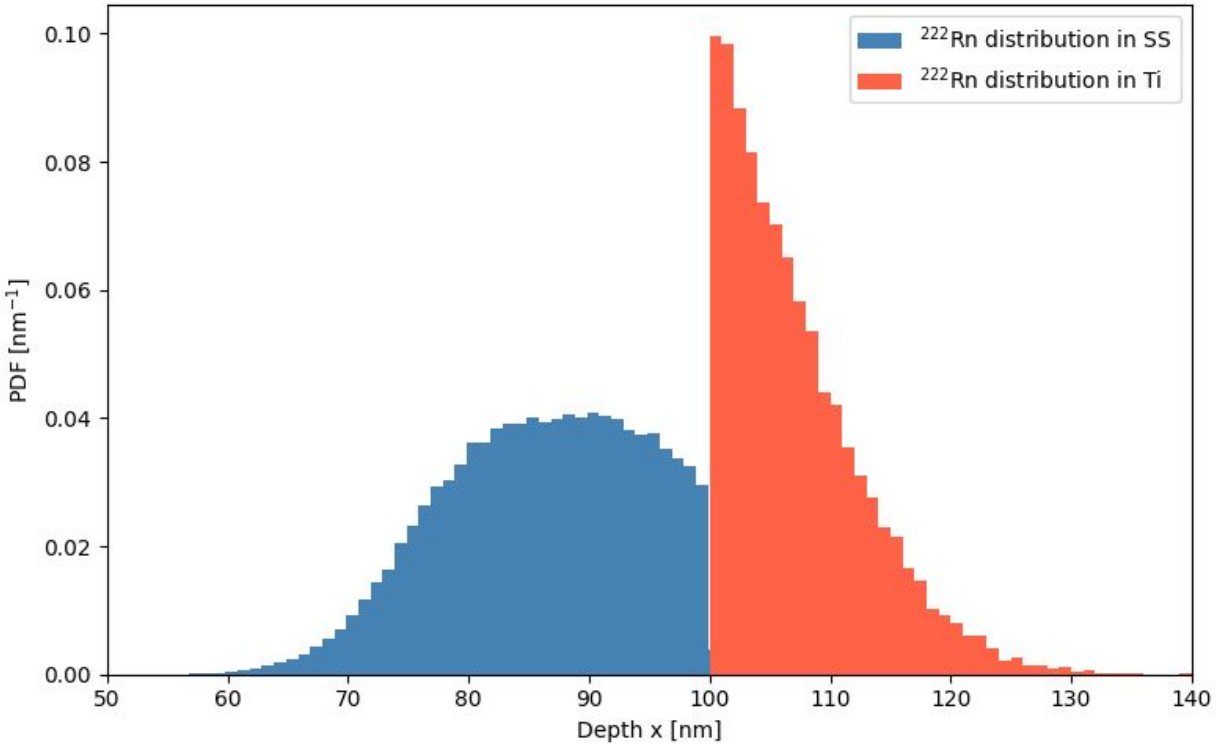
How?

- Put highest ^{226}Ra implanted source (SS new #05, ~ 217 Bq) in front of a “clean” sample, Rn will get implanted (17%) due to recoil from ^{226}Ra decay
- Use Cryogenic Radon Monitor to measure the emanated radon

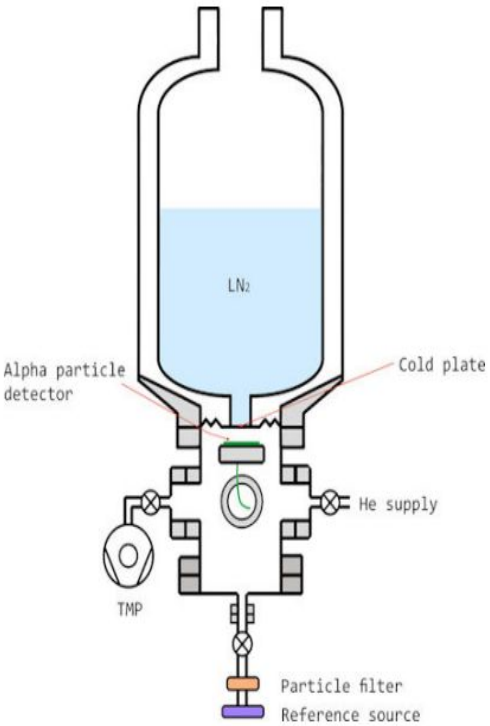


^{222}Rn diffusion coefficient measurement in solids: simulations

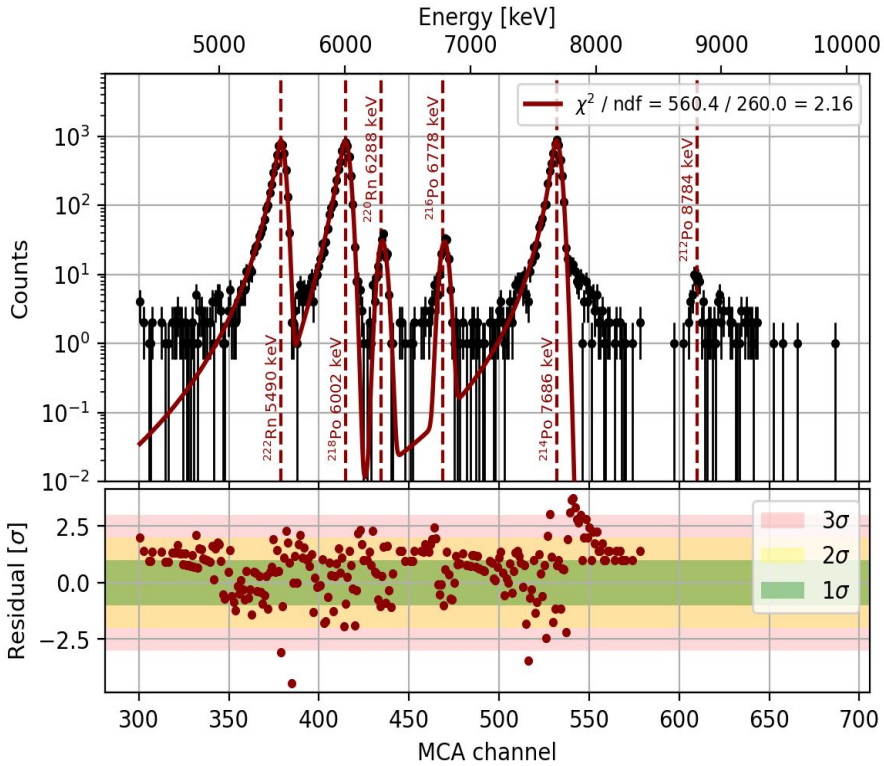
- ^{226}Ra source, Ra implanted in SS plate at ~11 nm depth
- Rn produced via α-recoil of ^{226}Ra with 86 keV kinetic energy
- SRIM simulation gives the Rn implantation depth distribution in the sample
- Sample placed 3 mm from SS source in vacuum, recoiling Rn implanted directly into material surface



The Cryogenic Radon Monitor



- Chamber split in two by a cold plate: liquid nitrogen (≈77 K) cools a Si PIN diode below it
- Lower volume evacuated (≈ 10^{-5} mbar): Rn diffuses in, freezes onto the plate/diode, and is detected upon decay
- Rn is measured directly, not its daughters → signal is an accumulation, not a decay
- 20 μBq minimum detectable ^{222}Rn activity



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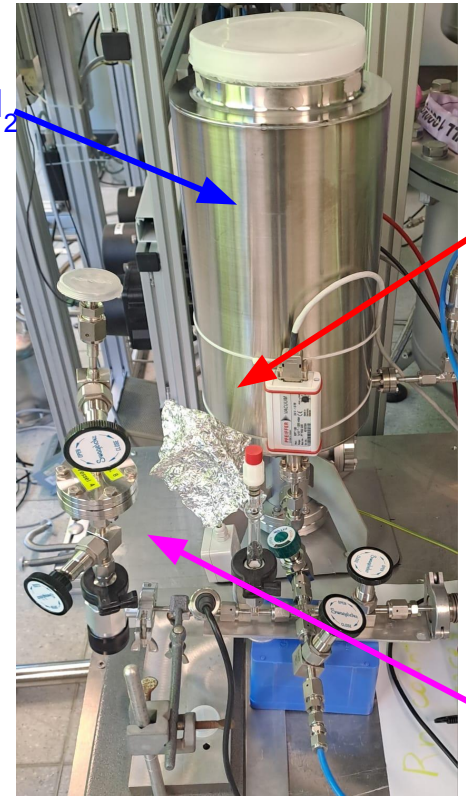

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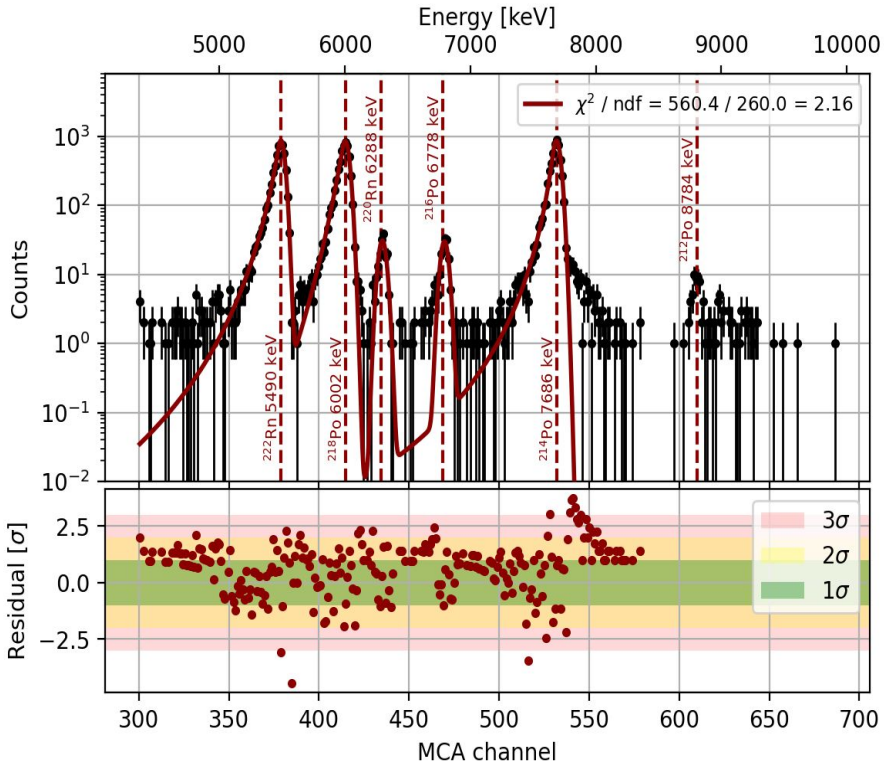
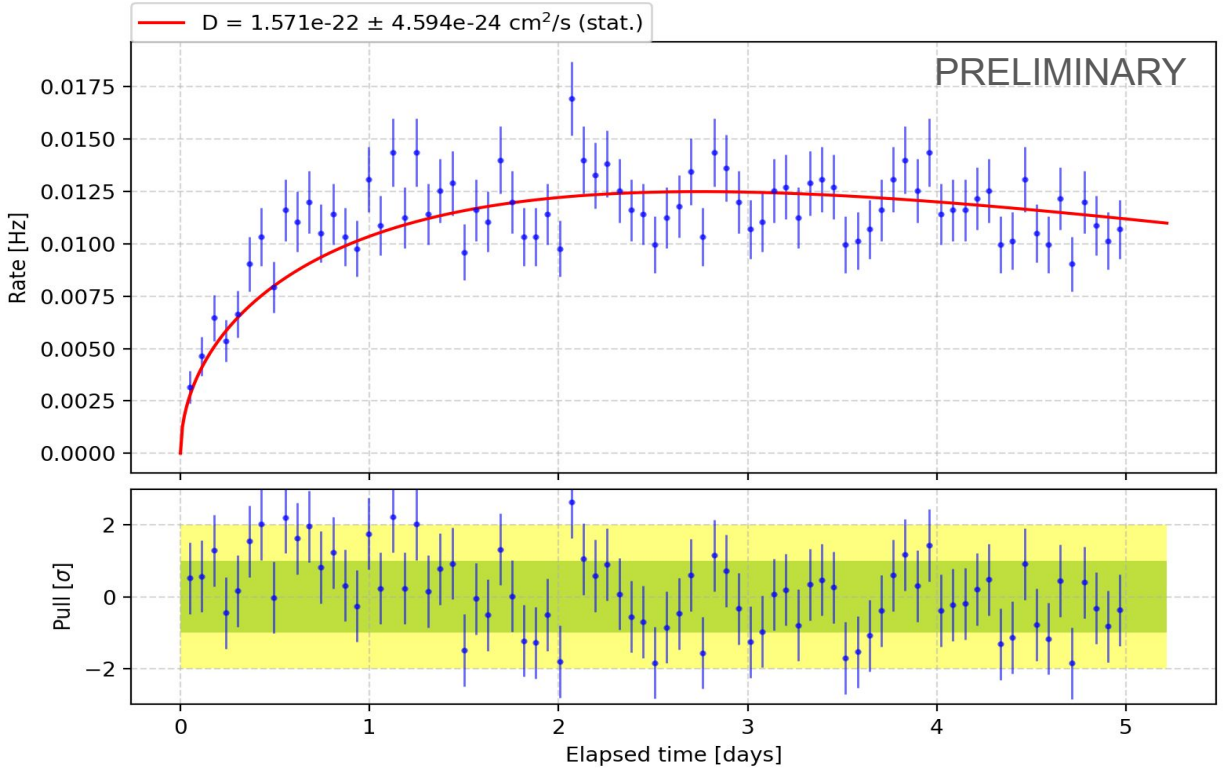


^{222}Rn diffusion coefficient measurement: example of copper

- Diffusion equation + Fick's law $J(t) = -D\,\partial_x C|_{x=0}$ determine ^{222}Rn evolution
- Select ^{222}Rn alpha peak
- Continuous rate measurement with the cryogenic radon monitor
- Fit to the outgassing rate model
- D ~ 10^{-22}–10^{-24} cm^2/s across Cu, Ti, stainless steel



Conclusions and outlook

Conclusions:

- Characterized 19 ^{226}Ra sources across 9 materials, validating TRIDYN against alpha/gamma spectroscopy and Rn emanation
- Recoil-driven emanation confirmed for SS, Ti, Cu, Si, quartz, Ge; diffusion dominates for PTFE, PEN, acrylic
- New α-recoil implantation technique: preliminary results for ^{222}Rn diffusion coefficient D ~ 10^{-22}–10^{-24} cm^2/s in Cu, Ti, and stainless steel

Next steps:

- ^{226}Ra implanted sources ready to be coated
- Assess reproducibility of Rn diffusion coefficient measurement
- Two implanted Si sources to be operated as bolometers for SURFαCE
- Paper in preparation

Thank you for your attention!

Lorenzo Ascenzo

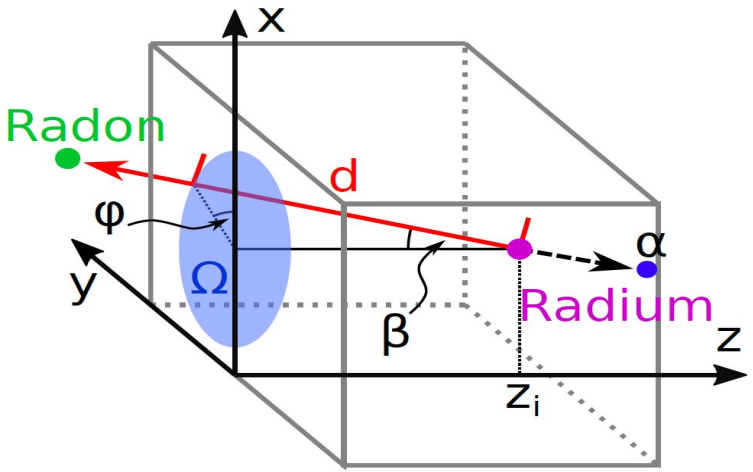
BACKUP slides

Lorenzo Ascenzo

Radon release mechanism: emanation by recoil

Emanation by recoil:

- α decay: $^{226}Ra \rightarrow ^{222}Rn + \alpha$
- 86 keV recoil energy → ~ 14nm SS, 50 nm Si



- To escape the sample, a radon nucleus formed at depth z_i must satisfy:

  $d = z_i / \cos(\theta) \leq R \rightarrow \cos(\theta\_crit) = z_i / R$

- Due to spherical symmetry, the escape solid angle is: **$\Omega = 2\pi\,(1 - \cos(\theta\_crit))$**

$$F = \frac{\Omega}{4\pi} = \frac{1}{2} \cdot \left(1 - \cos\left(\beta_{crit}\right)\right)$$

$$F = \frac{1}{2}\left(1 - \frac{z_i}{R}\right), \quad \text{for} \quad z_i \leq R$$

Emanation fraction of ^{222}Rn due to recoil: gaussian model

$$F = \frac{1}{A \cdot B} \cdot \int_0^\infty \int_0^\infty dr'\, dz'\, p_i(z') \cdot p_r(r') \cdot \begin{cases} \frac{1 - z'/r'}{2} & , \text{ for } z' \leq r' \\ 0 & , \text{ else} \end{cases}$$

Normalization constants

^{226}Ra implantation distribution

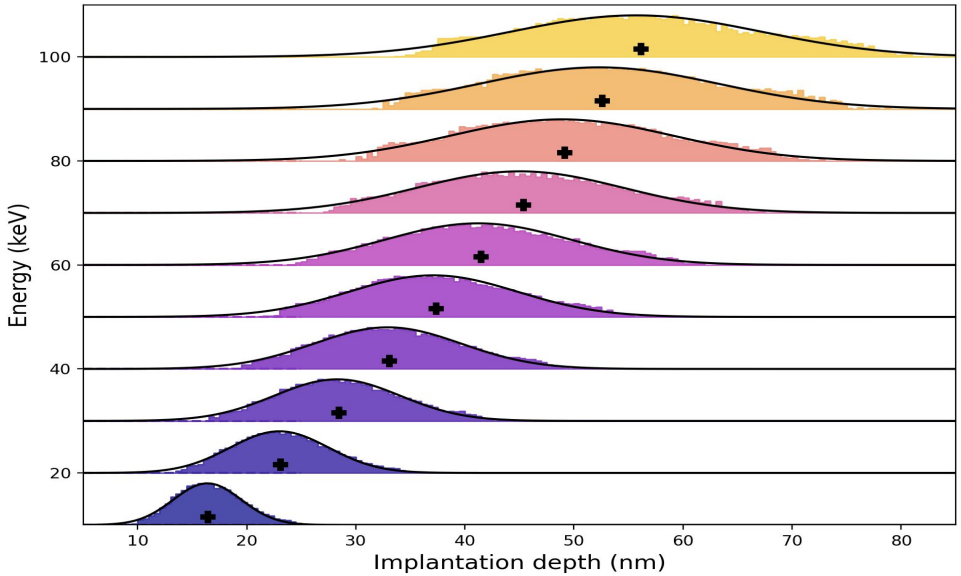
^{222}Rn recoil range

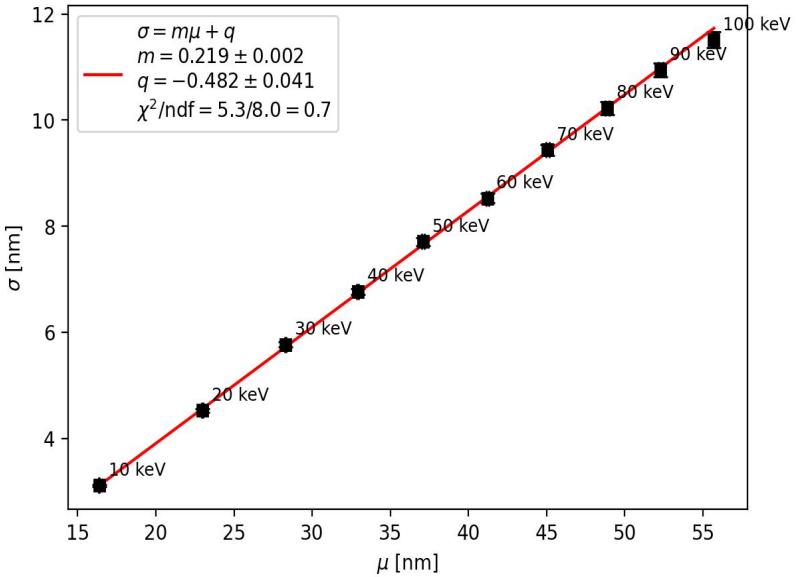
geometrical model

- The condition z'≤ r' is introduced to prevent the emanation fraction to become nonphysical and negative.
- ^{222}Rn recoil range distribution is fixed since ^{222}Rn recoil has fixed energy from α decay of ^{226}Ra
- ^{226}Ra distribution depends on the energy of implantation, to study the emanation fraction as a function of the Radium depth we need to perform simulations at different energies

Emanation fraction of ^{222}Rn due to recoil: gaussian model

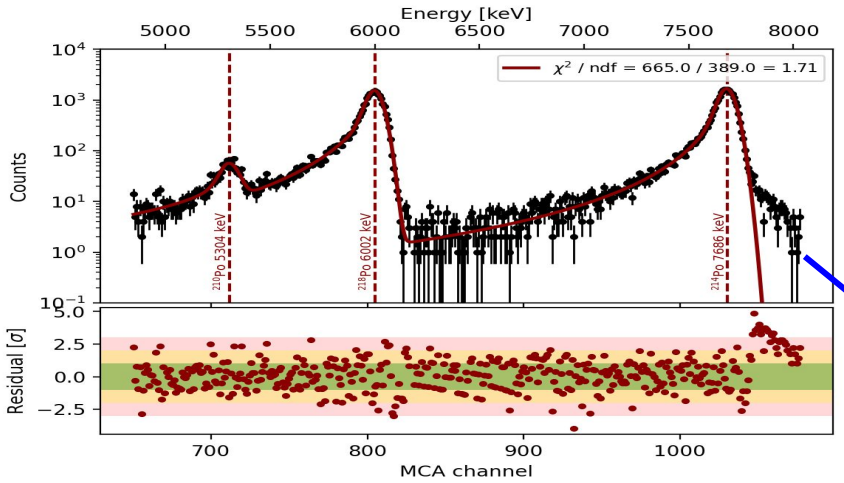
- Different implantation depths obtained by scanning ^{226}Ra energy from 10 to 100 keV
- Key observation: σ vs μ at different energies follows a **linear relationship** for all materials → allows generalization across the full energy range, obtain σ(μ)

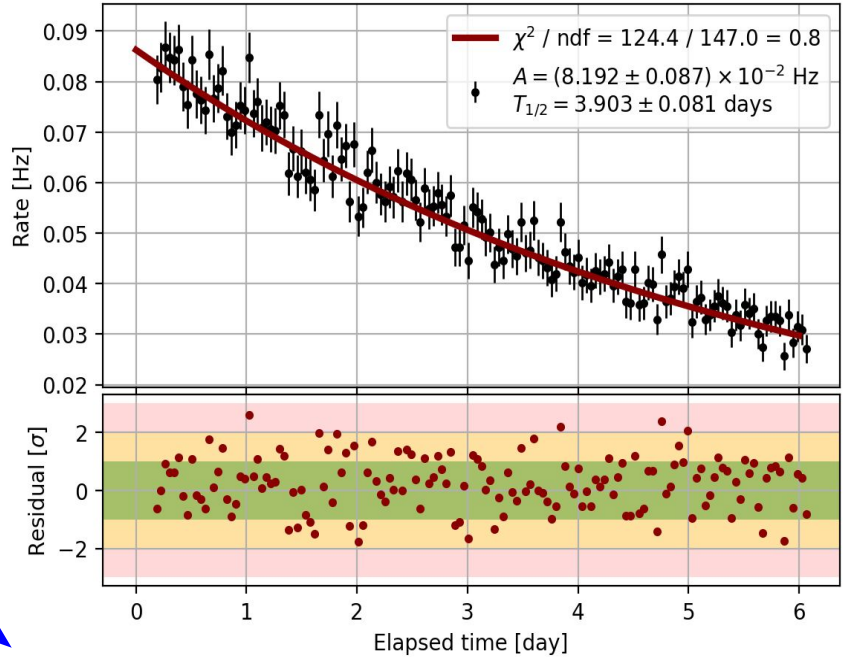




^{222}Rn emanation measurements: Electrostatic Radon Monitor

- ^{222}Rn activity extracted by fitting the **time evolution of ^{214}Po count rate** with an exponential decay (τ = ^{222}Rn lifetime)
- Background: ^{210}Po overlap with ^{218}Po → analysis restricted to the ^{214}Po energy range only
- Detection efficiency calibrated with a reference ^{226}Ra source (A = 21.54 ± 0.55 mBq): **ε = 28 ± 2%**





BiPo's

Evidence for diffusion in PTFE and PEN

- PTFE and PEN show emanation fractions close to 100%, well above the recoil-driven prediction (≈ 17%)

- Minimum diffusion length L_{min} inferred from the emanation fraction:
  PTFE #2: $L_{min} \approx 167$ nm $\rightarrow D_{min} \approx 5.8 \times 10^{-16}$ cm^2/s
  PEN #2: $L_{min} \approx 522$ nm $\rightarrow D_{min} \approx 5.7 \times 10^{-15}$ cm^2/s

- In both cases L_{min} exceeds the implantation depth by more than an order of magnitude

Cryogenic Radon Monitor calibration

- Reference sample: ISO-3 known ^{222}Rn emanation activity A_{ISO-3} = 1.976 ± 0.090 Bq
- Wrapped in PTFE foil: emanated Rn is stopped in the foil, then diffuses out, reproducing a pure-diffusion scenario
- Measured rate rises to saturation:

$$R(t) = A\left[1 - \exp\left(-\frac{t - t_0}{\tau}\right)\right]$$

with τ fixed to the known ^{222}Rn mean lifetime, A and t_0 free

- Efficiency from the fitted saturation rate A_∞:
  $\varepsilon = A_\infty / A_{ISO-3}$ = (35.0 ± 3.2) %