Production of ^{226}Ra-implanted high-quality radon sources for detector characterization

72nd Meeting of the INTC - 2023

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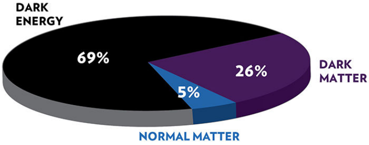
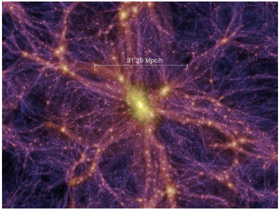
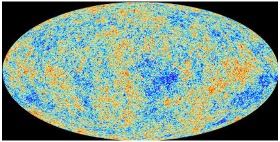
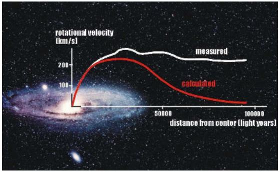
Many hints from Cosmology and Astronomy on its existence

Weakly interacting massive particles

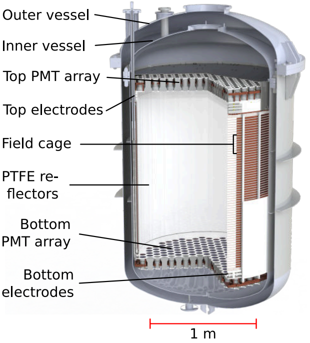




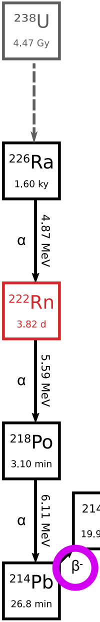


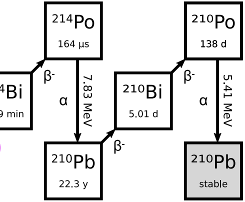


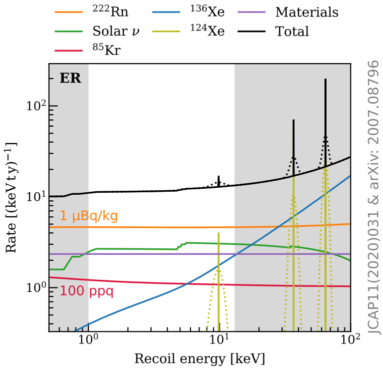
Direct search with liquid xenon detectors



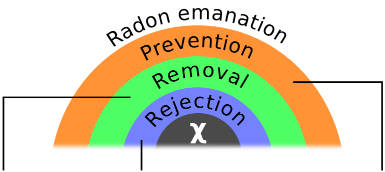
XENONnT Detector







- ^{222}Rn emanates from materials and mixes with LXe
- Dominant background in rare event searches
- Radon **mitigation** → radon **detection** → radon **sources**
- Also relevant in other fields (radioprotection, environmental physics, ...)!



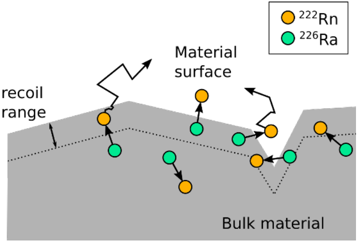
- Adsorption
- Distillation

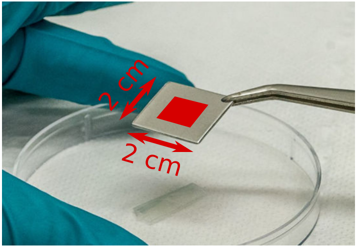
- ER/NR separation
- ...

- **Screening**
- **Coating**
- ...

New type of radon source: Proof-of-principle study

- Two samples with 5×10^{11} ^{226}Ra ions ($\approx 7\,$Bq) (2017)
- *Off-line* implantation using uranium carbide target (1.2×10^{18} POT)
- **G**eneral **p**urpose **s**eparator
- Ion beam (30 keV, 3pA), area of about $1 \times 1\,\mathrm{cm}^2$





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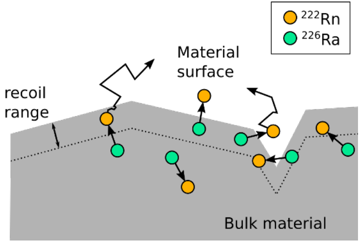
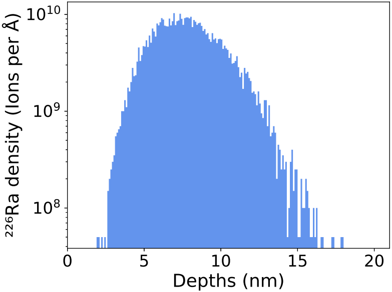


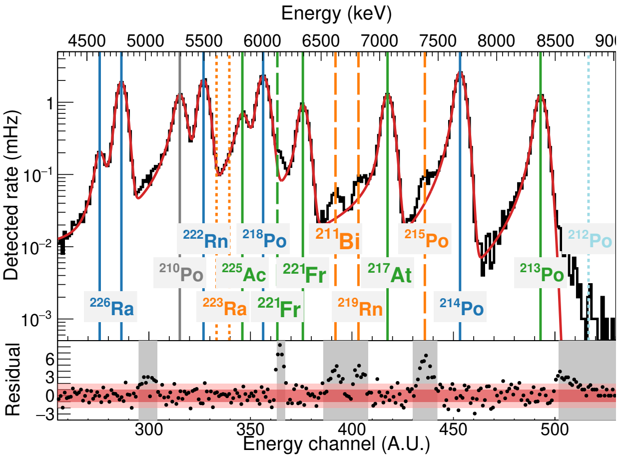
Simulated implantation profile

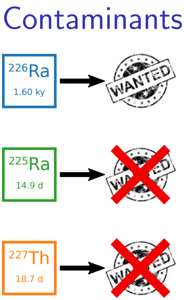
- Ion range distribution (SRIM) $\mu = 7.9\,\mathrm{nm}, \sigma = 2.3\,\mathrm{nm}$

+ High radon emanation fraction (expectation: 23%)
+ Mechanical stability assessed by wiping test: <1% removal



Observation of short-lived contaminants

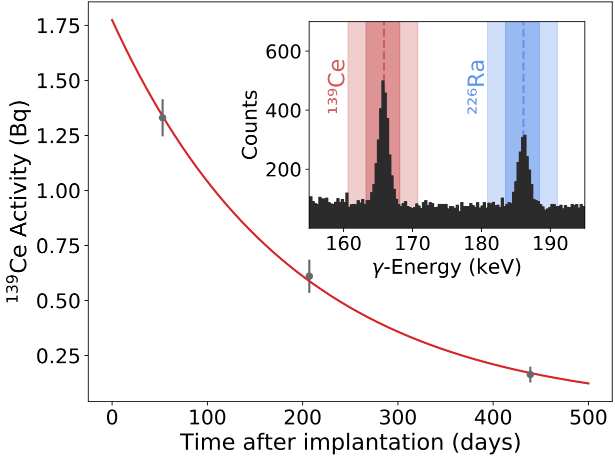




α-spectrum from 13 weeks after implantation:

- Unwanted isotopes decay with $\mathcal{O}(\text{weeks})$ of half-life
  $\rightarrow$ **no problem**

Unexpected ^{139}Ce contamination?

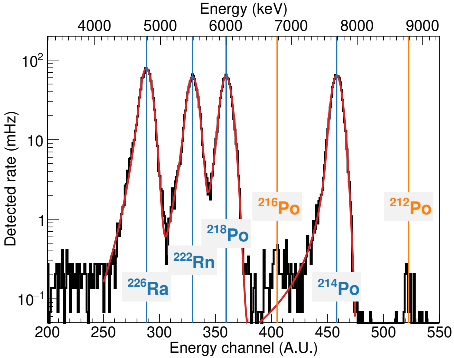


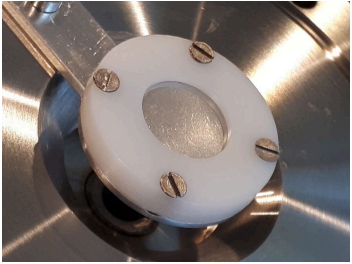
- Single γ-emission at 165.9 keV, matching ^{139}Ce
- Decay of this activity found with (130 ± 5) days, matching literature value of $T_{1/2}(^{139}\mathrm{Ce}) = 137.6\,\mathrm{days}$
- Present only on Sample B

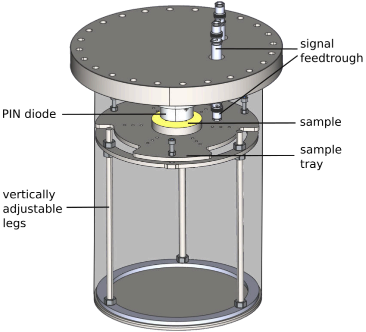
Alpha spectrometry (Si-PIN)

✓ Implantation confined within opening of holder ($1.9\,\mathrm{cm}$)

α-activity	^{226}Ra (4.9 MeV)
Sample A	$(8.7 \pm 1.9)\,\mathrm{Bq}$
Sample B	$(9.1 \pm 0.7)\,\mathrm{Bq}$



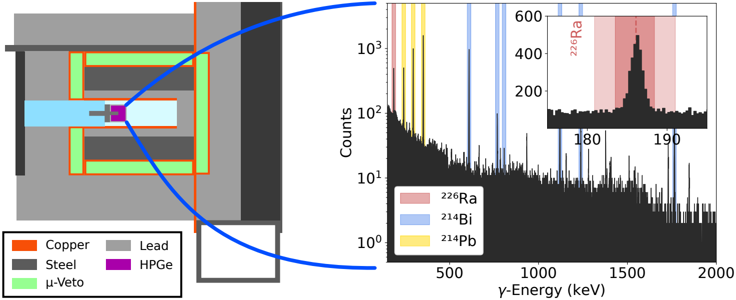




Eur. Phys. J. C 81, 343 (2021)

Gamma spectrometry (HPGe)

D. Budjáš PhD thesis (2009)



0.63 kg p-type HPGe crystal

- 15 mwe at MPIK, active & passive shielding

- N_2-flushed sample chamber
  $\Rightarrow$ emanated ^{222}Rn removed

Activity (Bq)	Sample A	Sample B
^{226}Ra (186 keV)	7.4 ± 0.9	8.4 ± 1.0
Rn-daughters	5.2 ± 0.3	6.0 ± 0.3

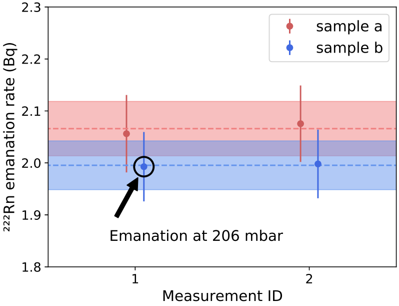
Emanation vessel

- Sample enclosed in vacuum tight stainless steel vessel
- Aluminum holder for fixation
- Vessel filled with helium (1050 mbar)

Results

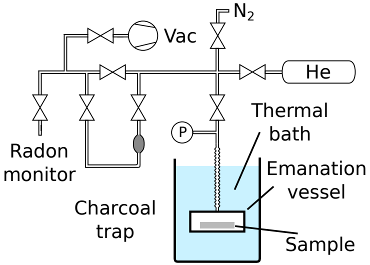
- Measured using low-level, quarz proportional counters
- Very good agreement

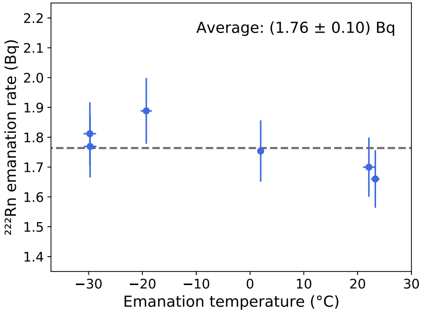
Sample	$^{222}\mathrm{Rn}$ emanation
Sample A	$(2.07 \pm 0.05)\,\mathrm{Bq}$
Sample B	$(2.00 \pm 0.05)\,\mathrm{Bq}$



Used setup

- Emanation vessel placed in thermal bath with 200 mbar Helium
- $^{222}\mathrm{Rn}$ emanation is collected on active charcoal trap ($\mathrm{LN_2}$ temperature)





Result

- $^{222}\mathrm{Rn}$ is then filled into electrostatic radon monitor
- No temperature variation down to $-30^{\circ}\mathrm{C}$

Results from full characterization

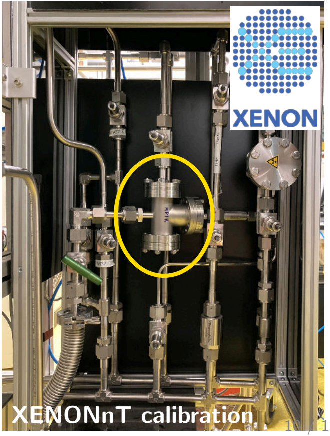
Activity (Bq)	Sample A	Sample B
Implantation	about 7	about 7
^{222}Rn emanation	$2.07 \pm 0.03_{\,\mathrm{stat}} \pm 0.04_{\,\mathrm{syst}}$	$2.00 \pm 0.03_{\,\mathrm{stat}} \pm 0.04_{\,\mathrm{syst}}$
γ-spectrometry	$7.4 \pm 0.1_{\,\mathrm{stat}} \pm 0.9_{\,\mathrm{syst}}$	$8.4 \pm 0.3_{\,\mathrm{stat}} \pm 1.0_{\,\mathrm{syst}}$
α-spectrometry	$8.70 \pm 0.06_{\,\mathrm{stat}} \,{}^{+2.0}_{-1.8}{}_{\,\mathrm{syst}}$	$9.13 \pm 0.10_{\,\mathrm{stat}} \,{}^{+0.7}_{-0.4}{}_{\,\mathrm{syst}}$

Appl.Radiat.Isot. 194 (2023) 110666 & arXiv: 2205.15926

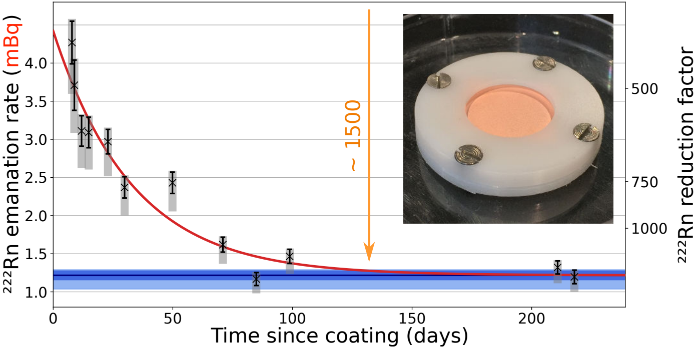
- Recoil dominated emanation of $^{222}\mathrm{Rn}$
  → Stable wrt. pressure, temperature, etc.
- Emanation from a bare stainless steel surface
  → Low outgassing of impurities

Applicability of implanted sources:

- Radon detectors and α-spectrometers
- Calibration of large scale LXe detectors!
- Radon mitigation studies (coating)



Results using ^{226}Ra implanted stainless steel samples



- ^{222}Rn reduction of ≈ 1500 using 5 µm electroplated copper
- Coating becomes “tighter” ⇒ room temperature annealing?

Activity (Bq)	Sample		Electrolyte
γ-Spectrometry	^{226}Ra	Rn-daughters	Rn-daughters
before coating	8.4±1.0	6.0±0.3	≲ 0.012
after coating	7.7±1.0	7.2±0.4	0.34±0.02

Proposal for ^{226}Ra implantation into further samples

- **Metals:** ~10x Stainless steel, 2x Copper, 2x Titanium
  Materials commonly used in low-background experiments (i.e. cryostats)
- **Insulators:**
  2x PTFE, 1x Quarz
  Light reflectors in LXe TPCs, Proportional counters
- **Semiconductors:**
  1x Si, 1x Ge
  Target materials for dark matter and $0\nu\beta\beta$ searches

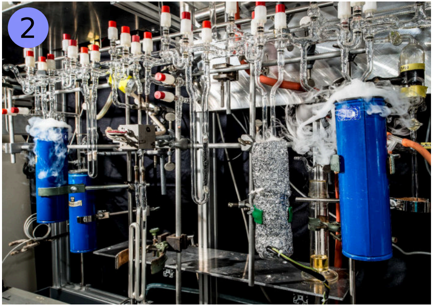
Isotope	Target	Ion source	Beam current
^{226}**Ra**	UC_x	Surface	3 pA

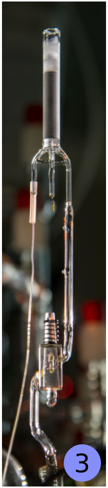
Samples	N_{ions}	Shifts	Protons required
20	3×10^{11}	12	**No**

  - Default material for coating studies: Stainless steel
  - Emanation fraction from diffusion vs. recoil in each material?
  - Influence of humidity and temperature?
  - Dependence on the gas type (He, N_2, Xe)?
  - Emanation into vacuum vs. gas vs. liquid?

Backup slides

1. Extraction of ^{222}Rn e.g. from ^{226}Ra solution
2. Transfer and purification of the sample
3. Counting (proportional counters, electrostatic radon monitors) MDA $\approx 30\,\mu$Bq





Appl. Radiat. Isot. 67, 5 889-893 (2009)

- Need reliable ^{222}Rn sources
- Radon mitigation using surface coatings

Bonus: Comparison to the PTB sample

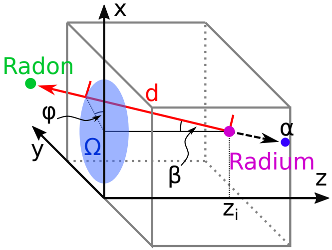
		MPIK	PTB
	Obtained samples	2×SS	4×Al + 4×W
Production	Implanted activity	about 7 Bq	0.5 kBq - 1 kBq
	Ion Energy	30 keV	30 keV
	^{226}Ra source	Produced in UC_x	^{226}Ra drop-cast on foil
	Ionization	Surface (3 pA)	Surface & Laser (10 nA)
Measurements	Emanation process	Recoil	Recoil
	Mechanical stab.	✓	✓
	Environment	cold, pressure	warm, humidity
	Reference	Appl. Radiat. Isot. **194**, 2023	Appl. Radiat. Isot. **181**, 2022

⇒ **Very promising source production process!**

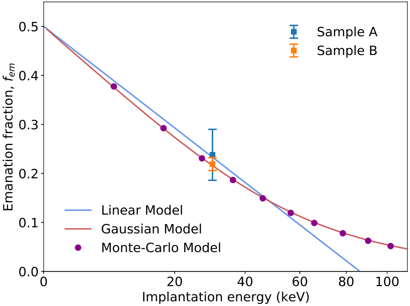
Linear model

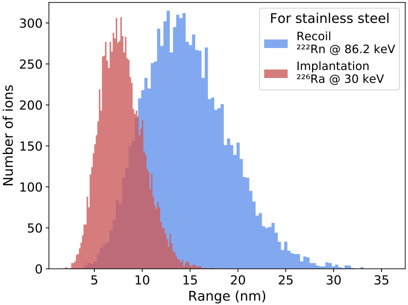
$$\Omega = \int_0^{2\pi} \int_0^{\beta_{crit}} \Big(\sin(\beta) \Big)\, d\beta\, d\varphi$$

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



Gaussian and Monte-Carlo model





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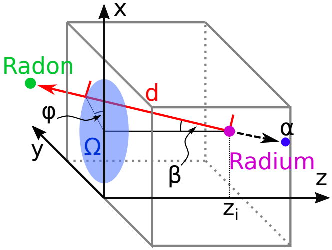

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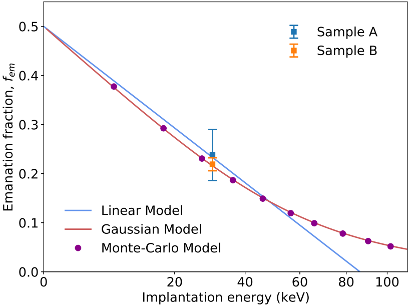
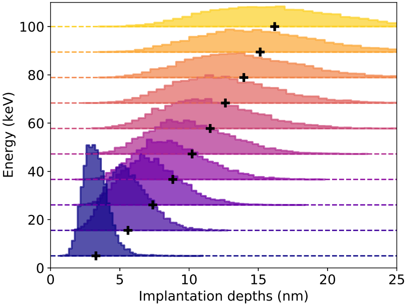
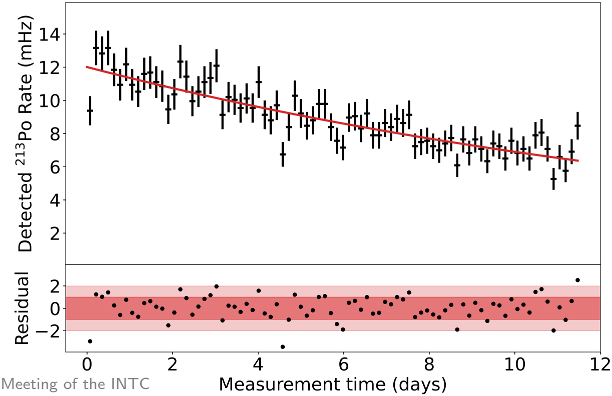




Short-lived isotopes details

Isotope	N_{ions}	Initial activity
^{226}Ra	10^{12}	$\mathcal{O}(10\,Bq)$
^{225}Ac	10^{9}	$\mathcal{O}(1\,kBq)$
^{227}Th	10^{7}	$\mathcal{O}(1\,Bq)$
$^{228}(Th/Ra)$	10^{6}	$\mathcal{O}(10\,mBq)$

$$^{225}Ra \xrightarrow[15\,d]{\beta} {}^{225}Ac \xRightarrow[10\,d]{5.9\,MeV} {}^{221}Fr \xRightarrow[4.9\,min]{6.5\,MeV} {}^{217}At \xRightarrow[32\,ms]{7.2\,MeV} {}^{213}Bi \xrightarrow[46\,min]{\beta} {}^{213}Po \xRightarrow[4.2\mu s]{8.5\,MeV} {}^{209}Pb$$



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$$^{227}\mathrm{Th} \xRightarrow[19\,\mathrm{d}]{6.0\,\mathrm{MeV}} {}^{223}\mathrm{Ra} \xRightarrow[11\,\mathrm{d}]{5.7\,\mathrm{MeV}} {}^{219}\mathrm{Rn}$$
$$\xRightarrow[4.0\,\mathrm{s}]{6.8\,\mathrm{MeV}} {}^{215}\mathrm{Po} \xRightarrow[1.8\,\mathrm{ms}]{7.4\,\mathrm{MeV}} {}^{211}\mathrm{Pb}$$
$$\xrightarrow[36\,\mathrm{min}]{\beta} {}^{211}\mathrm{Bi} \xRightarrow[2.1\,\mathrm{min}]{6.6\,\mathrm{MeV}} {}^{207}\mathrm{Tl}$$