The EMPIR initiative is co-funded by the European Union's Horizon 2020 research and innovation programme and the EMPIR Participating States

Development of integrated, primary ^{222}Rn emanation and measurement setup

EMPIR 19ENV01 traceRadon

This project 19ENV01 traceRadon has received funding from the EMPIR programme co-financed by the Participating States and from the European Union's Horizon 2020 research and innovation programme.

***19ENV01 traceRadon** denotes the EMPIR project reference.*

Florian Mertes

traceRadon scientific workshop, 20-10-2020

Preliminary remarks

- Stability of available ^{222}Rn emanation sources with respect to environmental parameters at least doubtful

  → Theoretically, dependency always exists
  → In-depth analysis of such dependencies is infeasible

- Demand for high precision at low ^{222}Rn emanation rates renders most techniques useless (e.g. γ-spectrometry)

  → Need for high efficiency detection of residual ^{222}Rn at low background (e.g. high S/N-ratio)

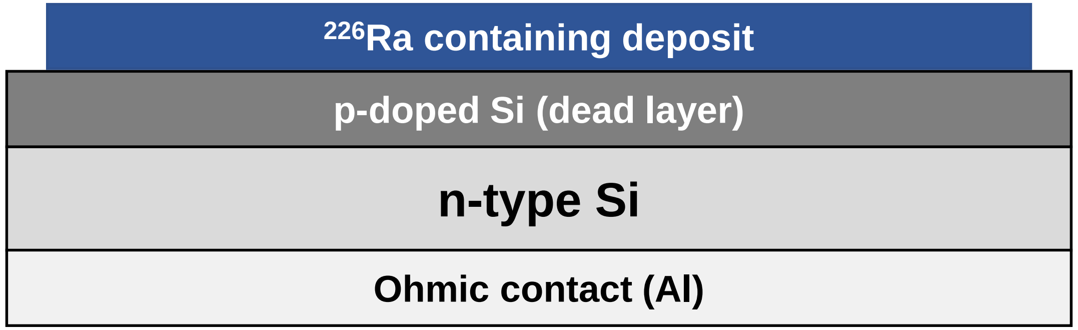
- Estimating ^{222}Rn emanation rate as a function of time from measuring residual ^{222}Rn is an inverse problem

General idea

$$^{226}\mathrm{Ra} \rightarrow {}^{222}\mathrm{Rn} \qquad + \qquad \alpha$$

E: 86 keV recoil
avg. range (Si): 40 nm [1]

E: 4.78 MeV
avg. range (Si): 23 µm [1]



Few µg / cm²
50 nm
> 100 µm

thin layer ^{226}Ra : recoil emanation of ^{222}Rn

close to 2π sr : ca. 50 % detection efficiency of all α

[1]: SRIM calculation

General idea

Benefits of this design:

- High efficiency to detect residual ^{222}Rn: Possibility to estimate emanation behaviour on-line and with comparably high temporal resolution (1000 s)

  → Possibly absolute measurements, to be investigated

- Low background

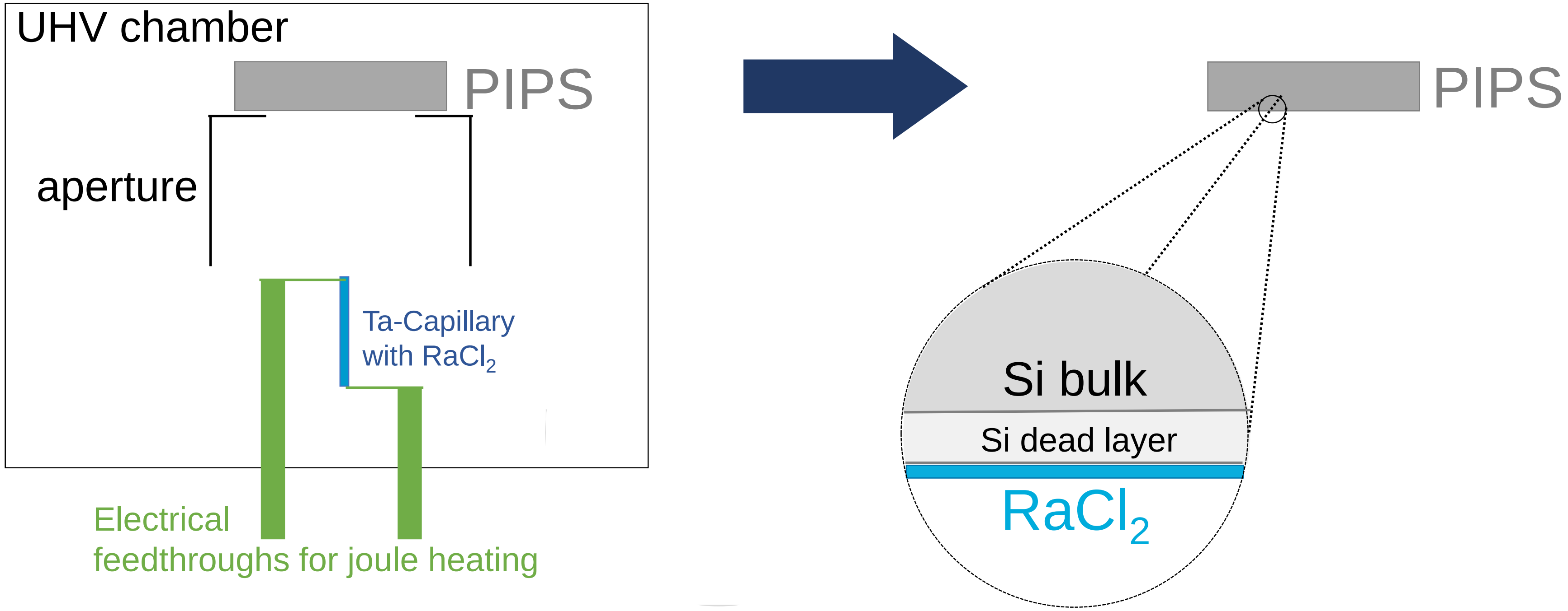
- Relatively rugged and cheap detectors (1 k€ for bare detector)

- Emanation due to recoil (rather than diffusive processes)
  → possibly diminished effect of environmental parameters

General idea

Challenge : Need ^{226}Ra-containing layer with low areal density

→ Modify commercial ion-implanted Si-detector with layer of $RaCl_2$ by thermal vapor deposition



UHV chamber

PIPS

aperture

Ta-Capillary with $RaCl_2$

Electrical feedthroughs for joule heating

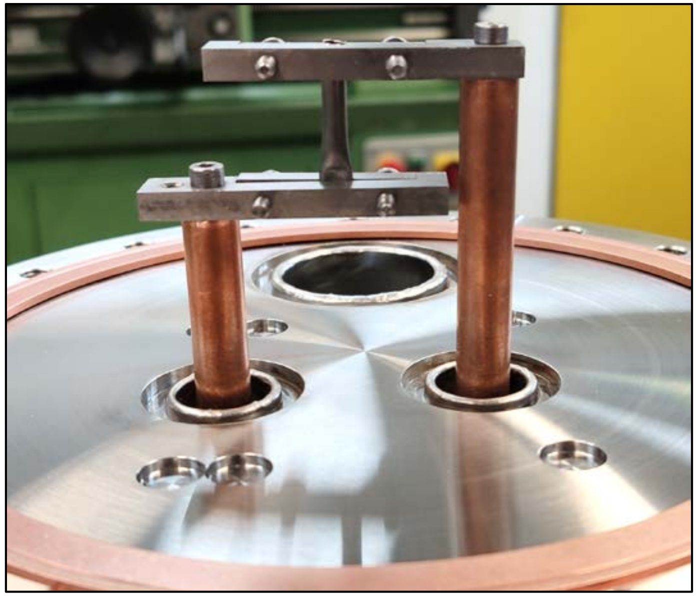
PIPS

Si bulk

Si dead layer

$RaCl_2$

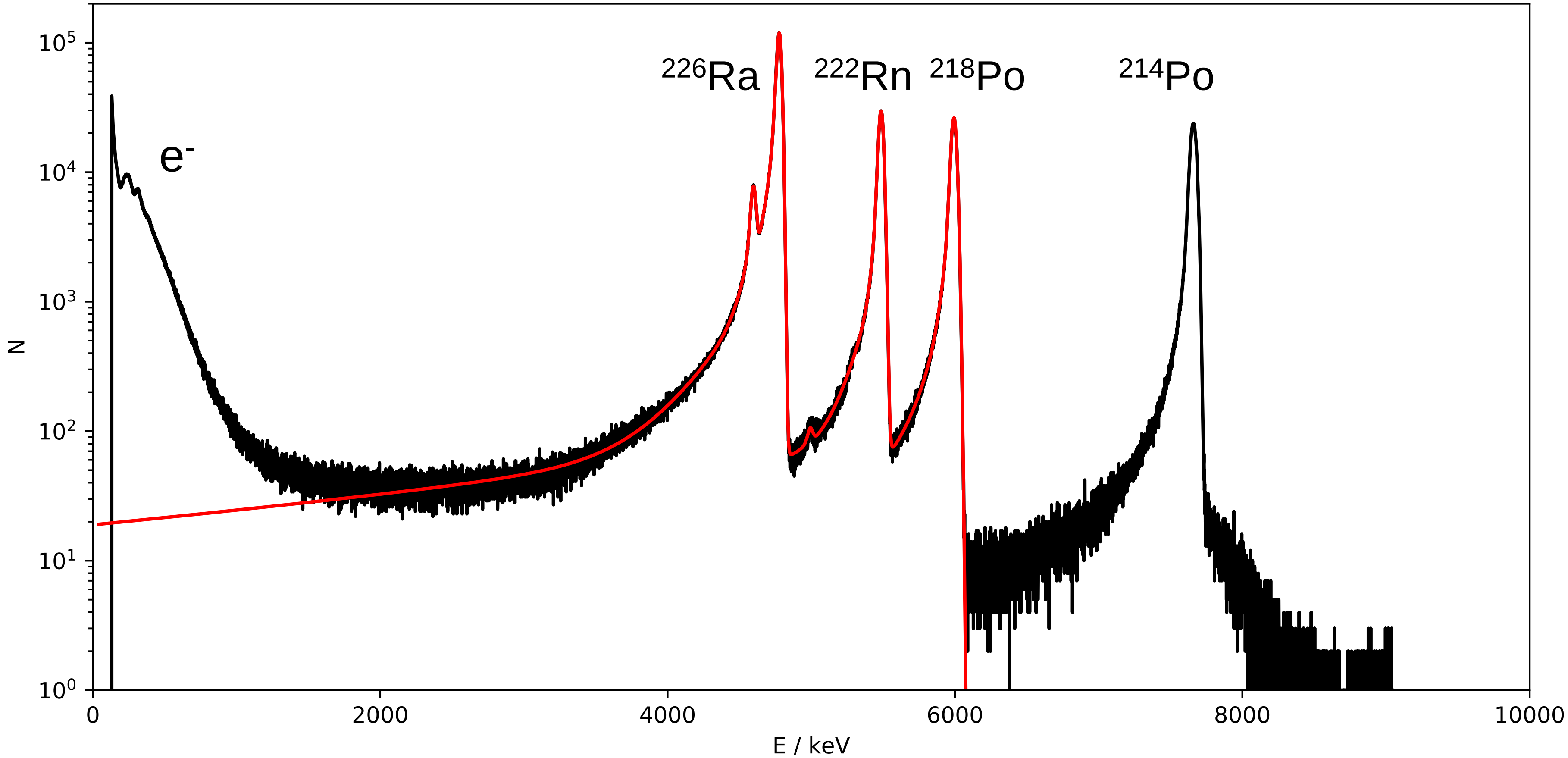
First evaporations and depositions



ca. 15 % yield (35 mm distance)
120 W, 15 min
$<10^{-6}$ hPa

PIPS 450 mm^2, 300 µm
with
150 Bq ^{226}Ra layer

First α spectra



Peaks are reasonably well resolved, still need to account for tailing contributions

→ Model with mixtures of Exponentially modified Gaussians

Estimating emanation rate

- generated ^{222}Rn atoms can either:
  - Escape the deposit due to recoil, diffusion ($\eta(t)$)
  - Stay within the deposit / get implanted into the dead-layer
- Spectral data allows to measure **residual** **^{222}Rn** (A_{Rn}^{S}) (and progeny) and ^{226}Ra (A_{Ra}^{S})

$$\mathit{First\ principles} \rightarrow \mathit{First\ order\ continuity}$$

$$\frac{dA_{Rn}^{S}}{dt} = -\lambda_{Rn}\, A_{Rn}^{S} + \lambda_{Rn}\, A_{Ra}^{S} - \lambda_{Rn}\, \eta(t)$$

Change = Decay + Ingrowth + Emanation

Estimating emanation rate

- Estimating the emanation rate $\eta(t)$ from time-series of spectra is thus an inverse-problem
- The time-series of A_{Rn}^{S} is the convolution of $\eta(t)$ with the impulse-response function of the system (given by DE)
- In the presence of measurement noise, **deconvolution** is generally **non-unique** and often numerically unstable
  → Statistical treatment (Bayes filters)
- **In closed systems**: steady-state, volumetric activity concentration of ^{222}Rn can be calculated more easily

Plans for the future

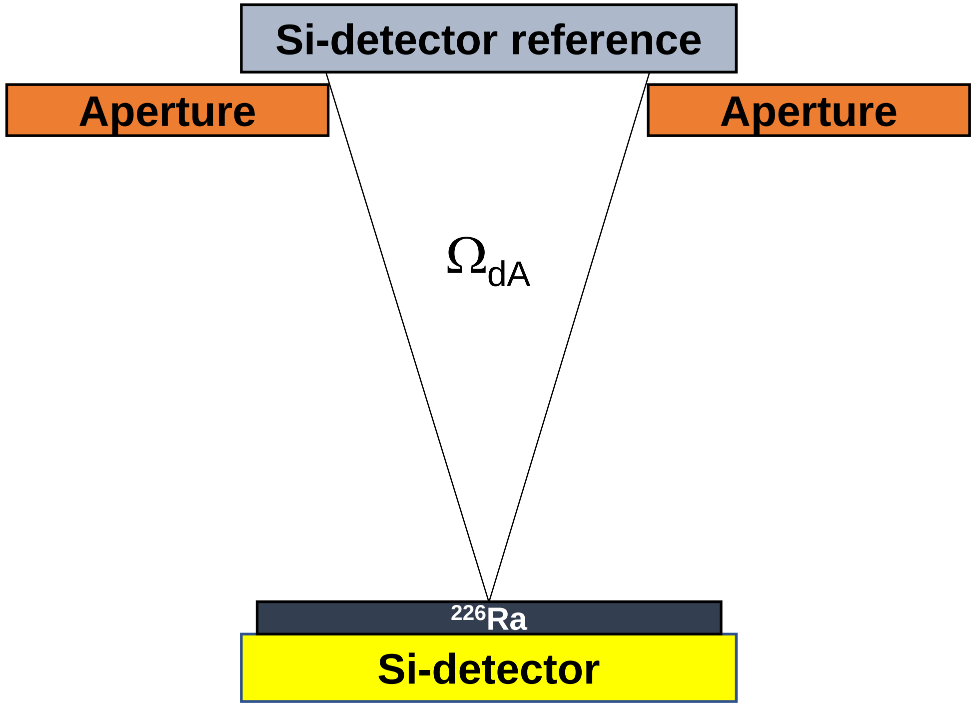
- Provide traceability for the presented design: Measure „against“ the primary, defined-solid angle α-particle spectrometer
- Produce several more systems of similar design
- Investigate long-term stability, stability with respect to environmental parameters, suitability for in-field calibration
- Improve and fine-tune algorithms for on-line operation of source

Questions?

florian.mertes@ptb.de

Backup # 1 : Traceability

Determination of detection efficiency by comparison with defined-solid angle spectrometer



Detection efficiency of reference detector is given by defined solid-angle (0.5 % rel. SD)

Calculated from

$$\epsilon = \frac{1}{4\pi} \frac{\int_A \Omega_{dA} w_{dA} dA}{\int_A w_{dA} dA}$$

Backup # 2 : Emanation rate

$$\frac{dA^S_{Rn}}{dt} = -\lambda_{Rn} A^S_{Rn} + \lambda_{Rn} A^S_{Ra} - \lambda_{Rn}\eta(t)$$

If we define

$$x = \begin{bmatrix} A^S_{Rn} \\ A^S_{Ra} \\ \eta(t) \\ \frac{d\eta(t)}{dt} \end{bmatrix} \quad F = \begin{bmatrix} -\lambda_{Rn} & \lambda_{Rn} & -\lambda_{Rn} & 0 \\ 0 & -\lambda_{Ra} & 0 & 0 \\ 0 & 0 & 0 & 1 \\ 0 & 0 & 0 & 0 \end{bmatrix} \quad L = \begin{bmatrix} 0 \\ 0 \\ 0 \\ 1 \end{bmatrix}$$

We can rewrite the system dynamics in terms of a vector valued stochastic differential equation (where $d\beta$ are increments of Brownian motion with spectral density Q)

$$dx = F dt + L d\beta$$

Backup # 2 : Emanation rate

$$dx = F dt + L d\beta$$

This system can be discretized for time-steps using the matrix-exponential function.

Given some (Gaussian) time-series of measurements and initial prior of components of x, the best-estimate (in terms of minimum variance) for this system is given by the Kalman-Filter and RTS-smoother.