First application of calorimetric low temperature detectors in accelerator mass spectrometry

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I. Motivation

Accelerator Mass Spectrometry (AMS)

= Standard method for the determination of very small isotope ratios

schematic overview:

In comparison with conventional mass spectrometry:

- background suppression due to use of an accelerator (e.g. breaking up molecules, separation of stable isobars)
- Sensitivity enhanced by about $10^5$

⇒ Determination of isotope ratios at a level of $10^{-10} - 10^{-16}$

→ Example: radio carbon dating: determination of $^{14}\text{C}/^{12}\text{C}$

Conventional semiconductor detectors/ionization chambers are limited in energy resolution, detection efficiency, resistivity against radiation damage etc.

⇒ Calorimetric low temperature detectors for heavy ions can provide an alternative.
II. Experimental Setup

a.) Calorimetric LTDs based on Al transition edge thermistors and sapphire absorbers

Thermistor: superconduction Al film

- Thin Al film ($d = 10$ nm): $T_C = 1.5$ K
  (photolithographically deposited onto sapphire substrate)
- Meander structure to match impedance to conventional preamplifiers
- Thermal coupling to heat sink via two gold wires ($\varnothing = 25$ $\mu$m)

Absorber: sapphire

- Dimensions: $330$ $\mu$m x $3$ mm x $2.5$ mm
- For sapphire: Debye temperature $\Theta_D = 1000$ K
- Single crystal material in high purity and with few lattice defects available
b.) AMS facility VERA = Vienna Environmental Research Accelerator

- Ions range from $^{10}$Be up to $^{238}$U
- Energy range 10 - 60 MeV
- Determination of isotope ratio:
  - stable isotope is measured as a current in a Faraday cup
  - radio isotope is detected in the heavy ion detector at the end of the beamline
III. Calorimetric LTDs in AMS

a.) Detector response to low energetic heavy ions


Systematic investigations for various ions ($^{12}$C, $^{197}$Au, $^{238}$U) at VERA:

achieved energy resolution
$\Delta E/E = 6 \times 10^{-3}$
especially for uranium
one order of magnitude
better than with conventional detectors

relative energy resolution:

no pulse height defect observed
b.) First application in an AMS experiment: precise determination of the isotope ratio $^{236}\text{U}/^{238}\text{U}$ in natural uranium

- **Motivation:**
  - $^{236}\text{U}$ produced by neutron capture:
    $$^{235}\text{U} + \text{n} \rightarrow ^{236}\text{U} + \gamma$$
  - Abundance of $^{236}\text{U}$ is an excellent neutron flux monitor for geological time scales (e.g. can prove the existence of so called „natural reactors“ like in Oklo, Gabon, West Africa)

- **Expected isotope ratio:**
  - in natural uranium: $^{236}\text{U}/^{238}\text{U} = 10^{-10} - 10^{-14}$
    - one of the heaviest radio nuclides interesting for AMS

- **First step:**
  - establish a material standard for which the absolute isotope ratio $^{236}\text{U}/^{238}\text{U}$ is precisely known.
  - increase sensitivity to measure down to $10^{-14}$

- **Experimental conditions:**
  - for detection of $^{236}\text{U}$: background from $^{234}\text{U}, ^{235}\text{U}, ^{238}\text{U}$
  - conventional setup: energy and TOF determination to distinguish between the different isotopes and suppress background
  - sensitivity limited due to detection efficiency in TOF setup (foils, scattering) and conventional detectors (entrance windows, dead layers)
IV. Results

a.) Detector performance

incident ion: $^{238}\text{U}$ at $E = 17.39$ MeV
operating point $T = 1.51$ K
temperature rise $\Delta T = 1.2$ mK

Preamplifier signal:

$\tau \approx 200 \mu\text{s}$

sufficient for typical countrates $\leq 10$ Hz

Spectrum: $\Delta E = 80$ keV, $\Delta E/E = 4.6 \times 10^{-3}$

(with conventional ionization chambers: $\Delta E/E \geq 5\%$)
Background suppression

Simulation: Separation of $^{234}\text{U}$, $^{235}\text{U}$, $^{236}\text{U}$, $^{238}\text{U}$ with $\Delta E/E = 4.6 \times 10^{-3}$:

assumed ratio $^{236}\text{U}$ to $^{234,235,238}\text{U} = 1:10$

⇒ advantages of calorimetric LTD:

- no TOF setup necessary
- no entrance foils

⇒ better detection efficiency
⇒ increased sensitivity
b.) Results of AMS measurements:

Investigated samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>Expected isotope ratio $^{236}\text{U}/^{238}\text{U}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>UD-150</td>
<td>$1.55 \times 10^{-9}$</td>
</tr>
<tr>
<td>UD-250</td>
<td>$2.58 \times 10^{-9}$</td>
</tr>
<tr>
<td>Joachimsthal 1</td>
<td>$10^{-11}$</td>
</tr>
<tr>
<td>Joachimsthal 2</td>
<td>$10^{-11}$</td>
</tr>
<tr>
<td>Bad Gastein</td>
<td>$10^{-12} - 10^{-14}$</td>
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</table>
Establishing a material standard: Samples from Joachimsthal

- Joachimsthal 1: $^{236}\text{U}/^{238}\text{U} = 4.35 \pm 0.09 \pm 0.69 \times 10^{-11}$
- Joachimsthal 2: $^{236}\text{U}/^{238}\text{U} = 2.65 \pm 0.08 \pm 0.42 \times 10^{-11}$

- Limiting factor: systematic error from determination of transmission $0.65 \pm 0.1$

- Result of earlier measurement with conventional E/TOF setup: transmission $0.2 \pm 0.06$, sample Joachimsthal 1: $^{236}\text{U}/^{238}\text{U} = 6.1 \pm 0.4 \pm 1.8 \times 10^{-11}$

(C. Vockenhuber et al., Int. J. Mass Spectrometry 223-224 (2003) 713)
Determination of $^{236}\text{U}/^{238}\text{U}$ in spring water from Bad Gastein

→ limited amount of sample material!

With 65% transmission and 30 minutes of measurement time:

$^{236}\text{U}/^{238}\text{U} = 1.2 \pm 0.3 \pm 0.2 \times 10^{-11}$

= smallest isotope ratio for $^{236}\text{U}/^{238}\text{U}$ ever measured

(Factor of 6 lower as compared to previous measurements)!

Limiting factor: statistics!
Conclusion

Calorimetric low temperature detectors for heavy ions

- show good energy resolution of $\Delta E/E = 4.6 \times 10^{-3}$ for low energetic heavy ions (one order of magnitude better than conventional detectors)
- are therefore suitable as replacement for a conventional E/TOF spectrometer in AMS
- substantially improve background discrimination and detection efficiency

A first AMS measurement to precisely determine the isotope ratio of $^{236}\text{U}/^{238}\text{U}$ in the AMS laboratory VERA resulted in

- a new material standard with higher precision
- improved level of sensitivity: $^{236}\text{U}/^{238}\text{U} = 1.2 \times 10^{-11}$ (smallest isotope ratio ever measured)

Perspectives

- energy resolution improved by factor of 2:
  $\Rightarrow$ better background suppression
  $\Rightarrow$ increase of sensitivity to measure smaller isotope ratios
- larger detector surface /detector array: increase transmission to 100 %
  $\Rightarrow$ systematic error from transmission eliminated
- investigation of Pu isotopes