

# First application of calorimetric low temperature detectors in accelerator mass spectrometry

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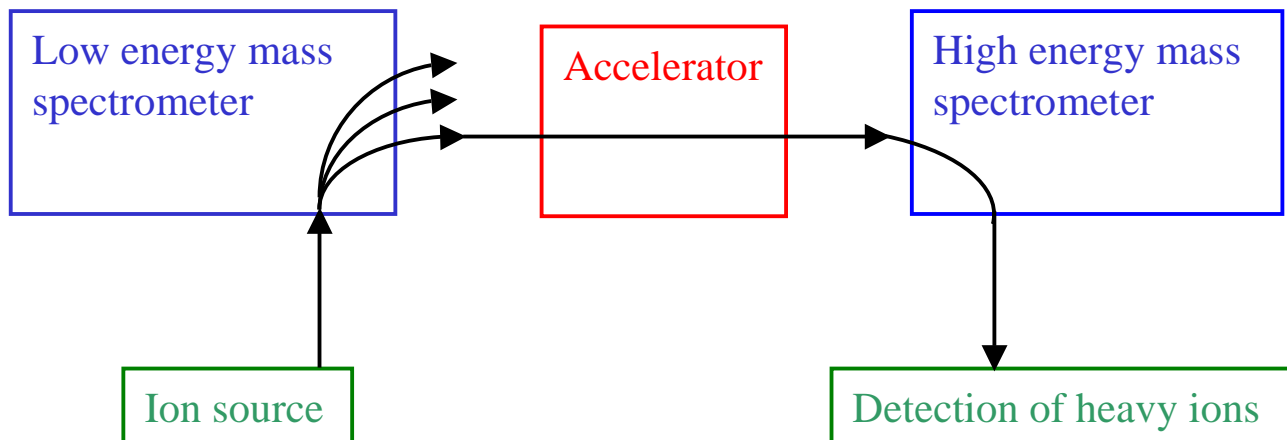


# I. Motivation

## 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.

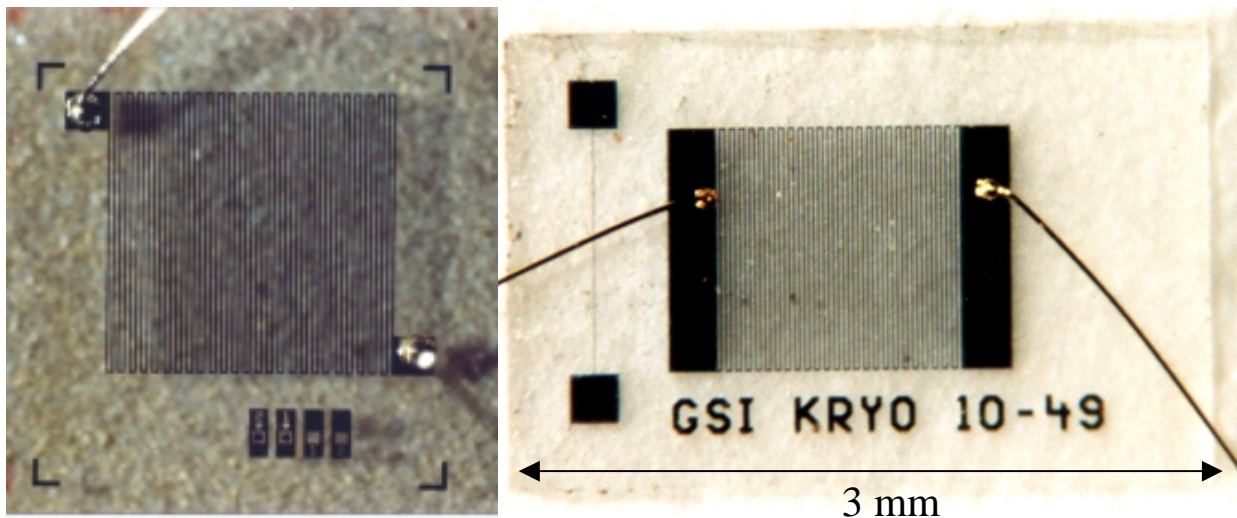
⇒ Calorimetric low temperature detectors for heavy ions can provide an alternative.

## II. Experimental Setup

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

#### Thermistor: superconduction Al film

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

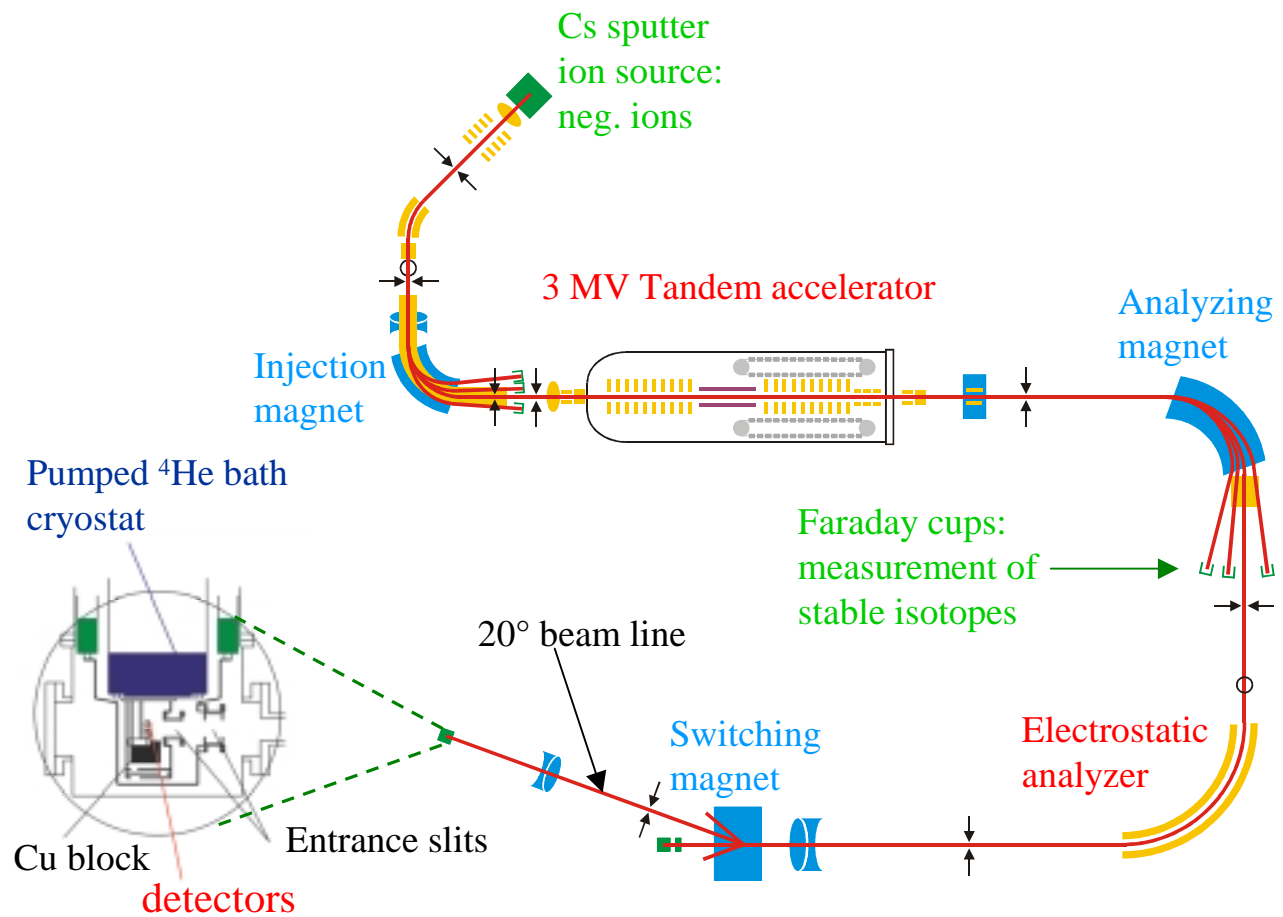


Al thermistor deposited onto sapphire substrate

#### Absorber: sapphire

- Dimensions:  $330 \mu\text{m} \times 3 \text{ mm} \times 2.5 \text{ mm}$
- For sapphire: Debye temperature  $\Theta_D = 1000 \text{ K}$
- Single crystal material in high purity and with few lattice defects available

## b.) AMS facility VERA = Vienna Environmental Research Accelerator



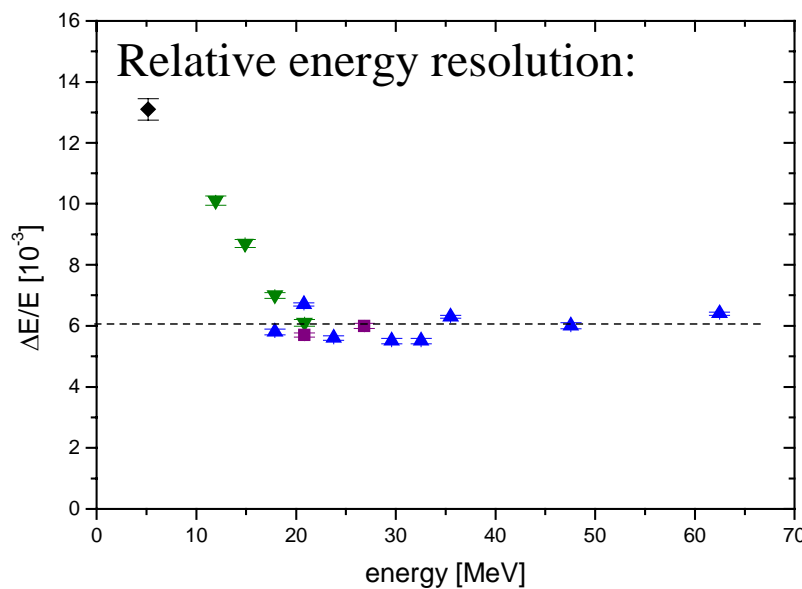
- Ions range from  $^{10}\text{Be}$  up to  $^{238}\text{U}$
- Energy range 10 - 60 MeV
- Determination of isotope ratio:
  - stable isotope is measured as a current in a Faraday cup
  - radio isotope is detected in the heavy ion detector at the end of the beamline

# III. Calorimetric LTDs in AMS

## a.) Detector response to low energetic heavy ions

(S. Kraft et al., AIP Conf. Proc. LTD-9 (2002) 405)

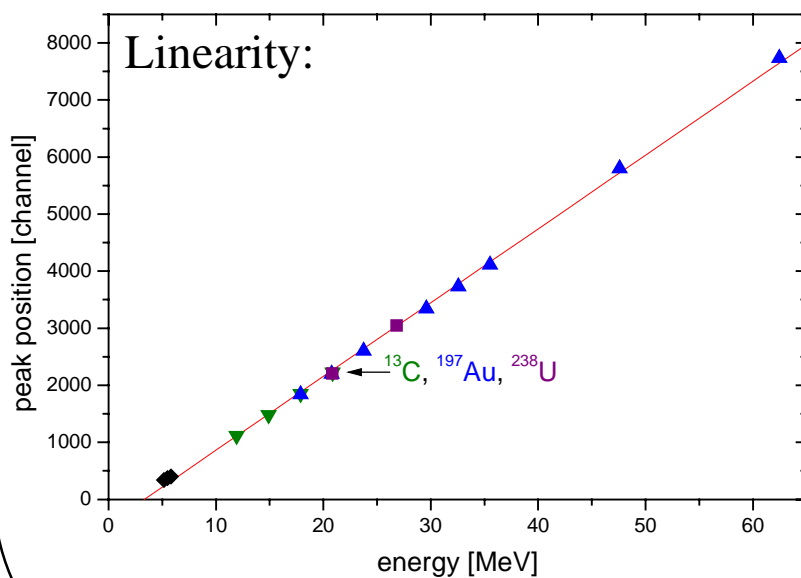
Systematic investigations for various ions ( $^{12}\text{C}$ ,  $^{197}\text{Au}$ ,  $^{238}\text{U}$ ) at VERA:



achieved energy resolution

$$\Delta E/E = 6 \times 10^{-3}$$

especially for uranium  
one order of magnitude  
better than with  
conventional detectors



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

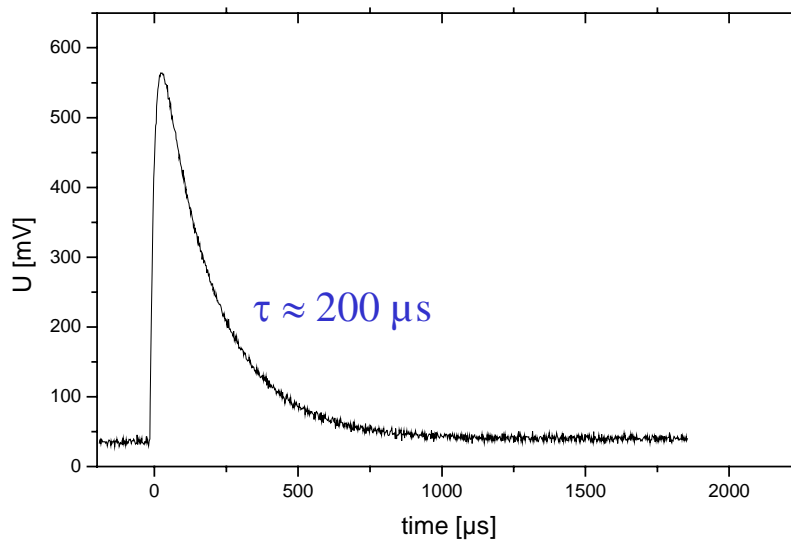
### a.) Detector performance

incident ion:  $^{238}\text{U}$  at  $E = 17.39 \text{ MeV}$

operating point  $T = 1.51 \text{ K}$

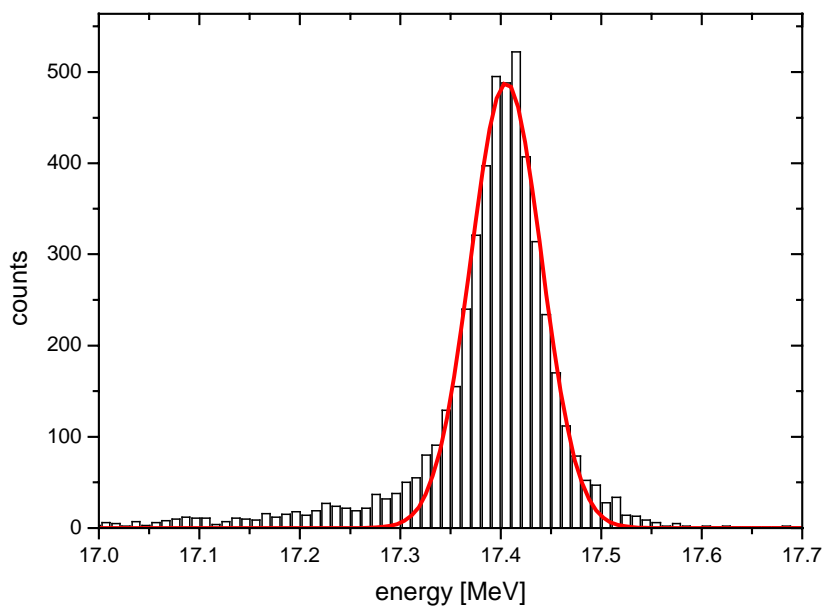
temperature rise  $\Delta T = 1.2 \text{ mK}$

Preamplifier signal:



→ sufficient  
for typical  
count rates  
 $\leq 10 \text{ Hz}$

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

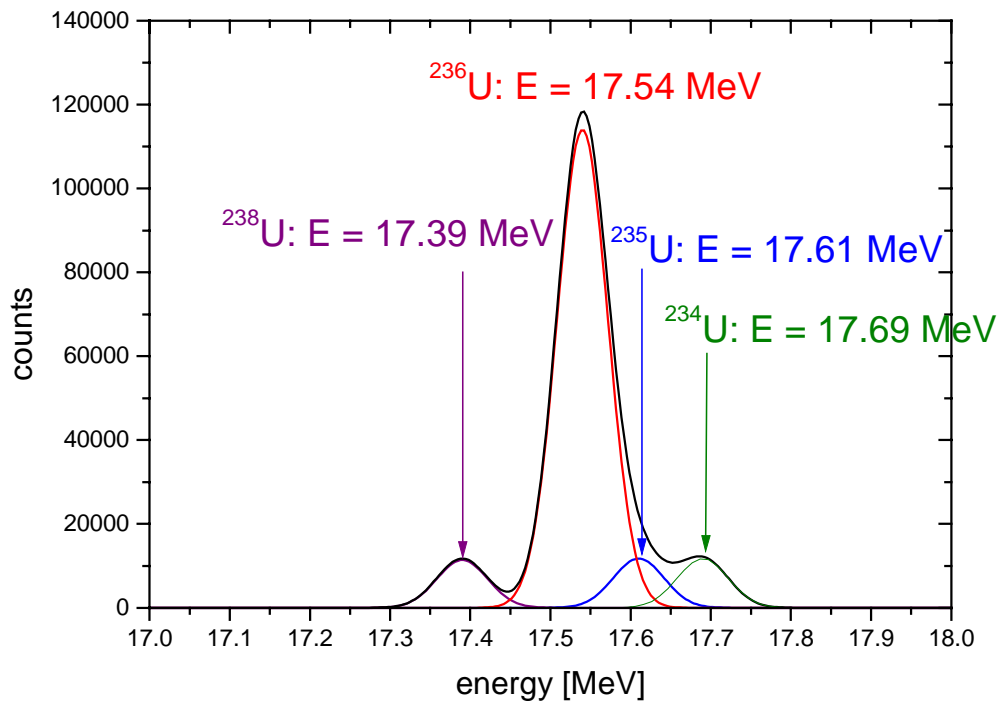


(with  
conventional  
ionization  
chambers:  
 $\Delta E/E \geq 5 \%$ )

## 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



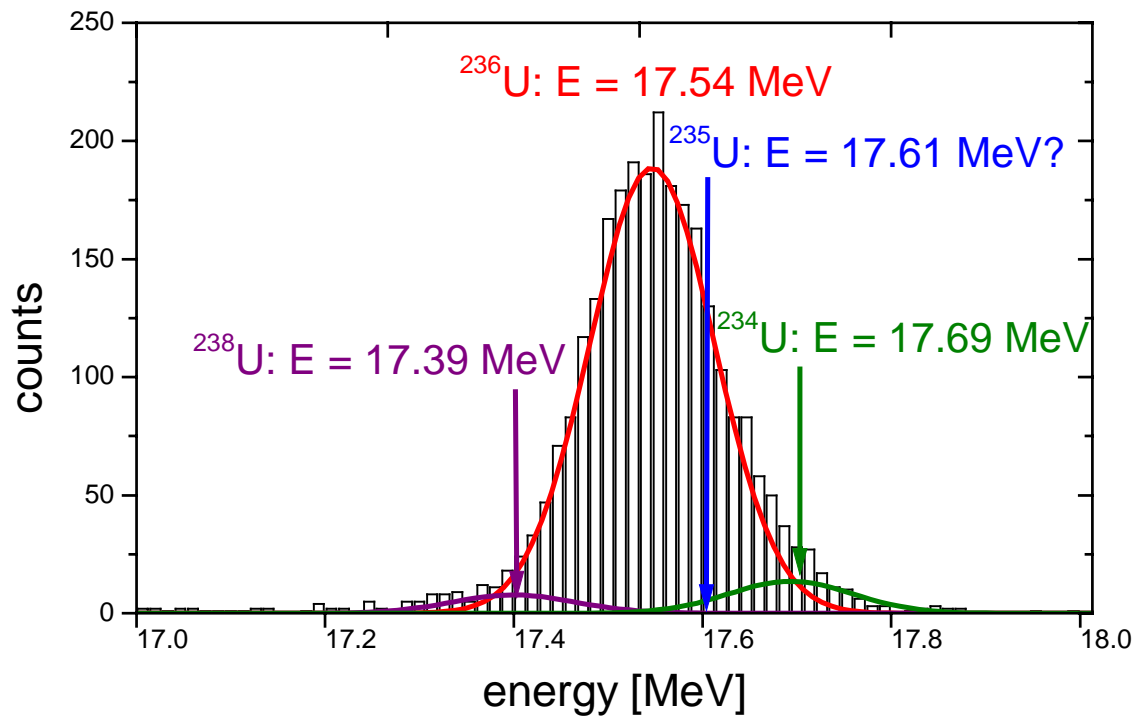
## b.) Results of AMS measurements:

### Investigated samples

Sample	Expected isotope ratio $^{236}\text{U}/^{238}\text{U}$
UD-150	$1.55 \times 10^{-9}$
UD-250	$2.58 \times 10^{-9}$
Joachimsthal 1	$10^{-11}$
Joachimsthal 2	$10^{-11}$
Bad Gastein	$10^{-12} - 10^{-14}$

# Establishing a material standard: Samples from Joachimsthal

Detector performance:  $\Delta E = 159 \text{ keV}$ ,  $\Delta E/E = 9.1 \times 10^{-3}$



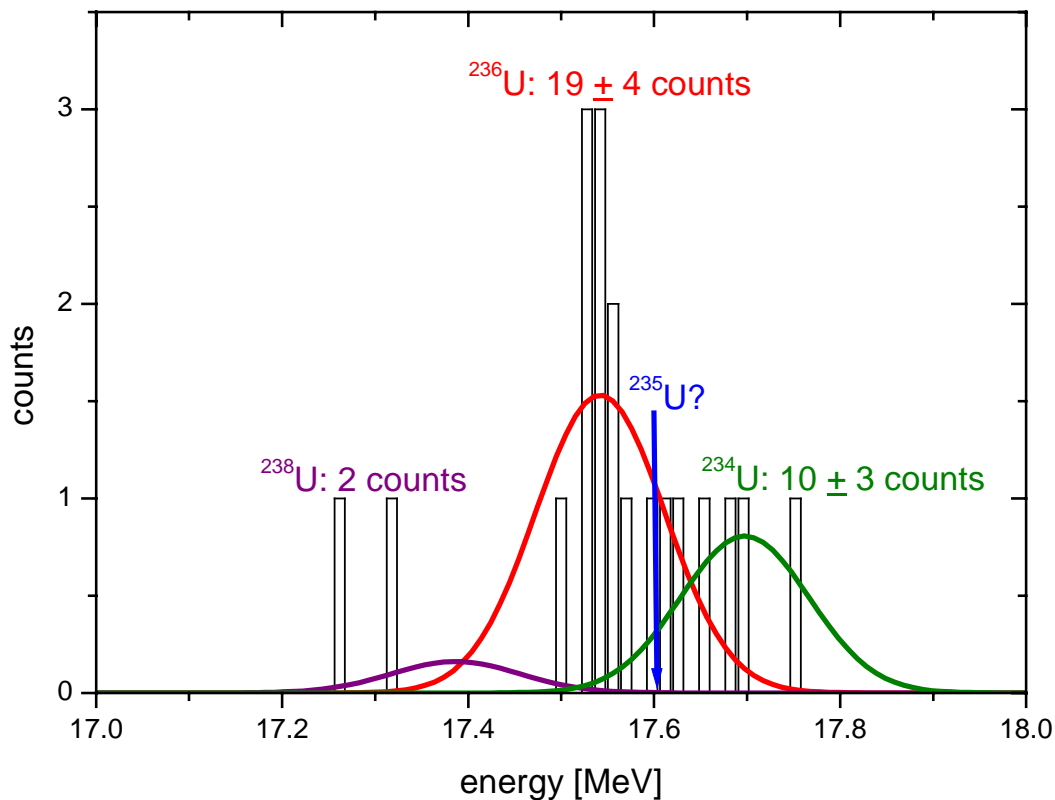
- Joachimsthal 1:  $^{236}\text{U}/^{238}\text{U} = 4.35 \pm 0.09 \pm 0.69 \times 10^{-11}$
- Joachimsthal 2:  $^{236}\text{U}/^{238}\text{U} = 2.65 \pm 0.08 \pm 0.42 \times 10^{-11}$
- Limiting factor: **systematic error** from determination of transmission  $0.65 \pm 0.1$
- Result of earlier measurement 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:
  - ⇒ better background suppression
  - ⇒ increase of sensitivity to measure smaller isotope ratios
- larger detector surface /detector array: **increase transmission to 100 %**
  - ⇒ systematic error from transmission eliminated
- **investigation of Pu isotopes**