AMS measurements of ^{55}Fe in steel

- an example of a simple analysis with a big machine

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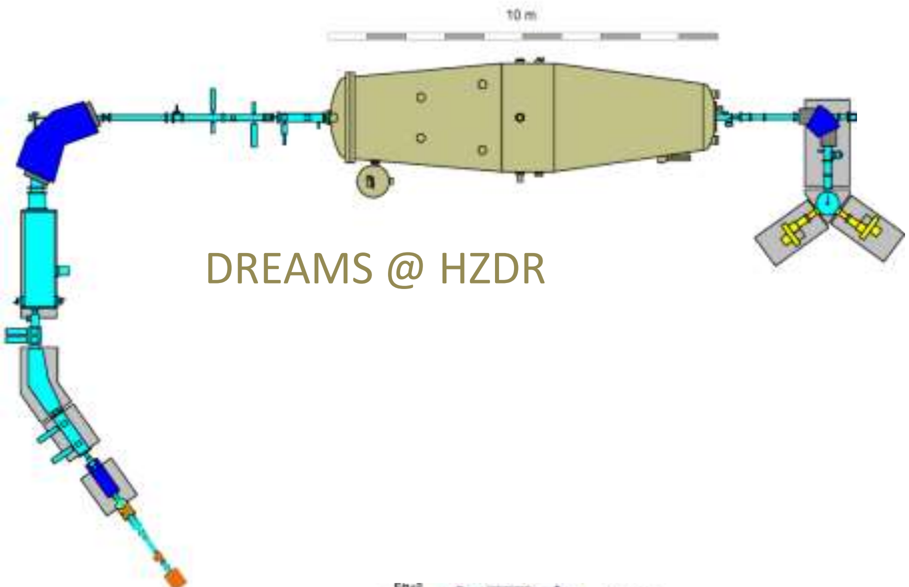
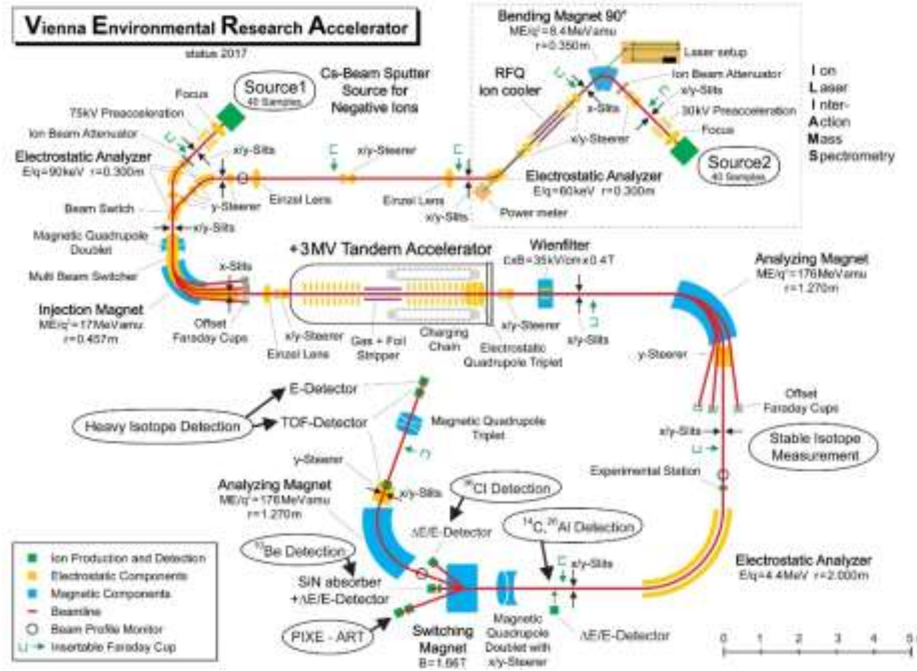
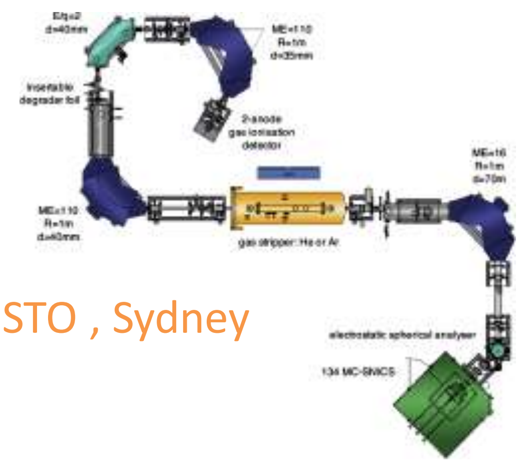
Accelerator Mass Spectrometry (AMS) instruments



DREAMS @ HZDR



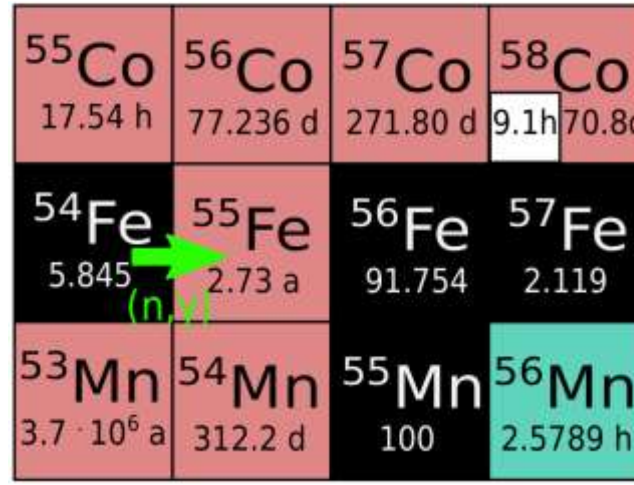


VEGA @ ANSTO , Sydney

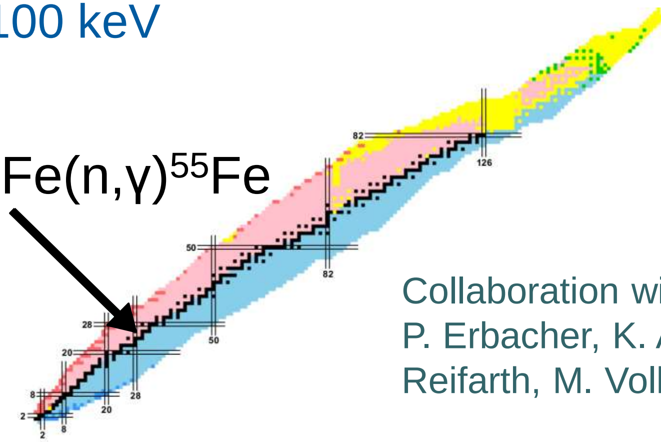
MICADAS @ ETH Zurich

Why ^{55}Fe?

1) Understanding stellar nucleosynthesis requires experimental cross sections at s-process-neutron energies: E_n ~ 1 – 100 keV



^{54}Fe(n,γ)^{55}Fe



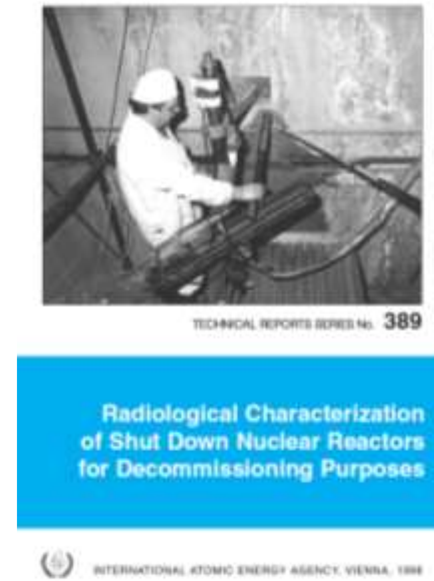
Collaboration with Z. Slavkovská, B. Brückner, P. Erbacher, K. Al-Khasawneh, S.Pavetich, R. Reifarth, M. Volknandt, M. Weigand

2) Production inside a nuclear reactor via ^{54}Fe(n,γ)^{55}Fe and ^{56}Fe(n,2n)^{55}Fe on steel components (reactor vessels, tubes, steel in concrete)
"short" half-life $T_{1/2}$=2.7563 ± 0.0004 years (Pommé et al., 2019)
→ high levels in first years after shutdown

Important & hard to measure radionuclides

Cited from IAEA Radiological Characterization of Shut Down Nuclear Reactors for

Decommissioning Purposes, Technical Reports Series No. 389, Vienna 1998



Fe 55
2.73 a
ε
no γ
σ 13
$\sigma_{n,\alpha}$ 0.01

"For GCRs and other reactors, **^{55}Fe** is the major short term component of the radioactive inventory following shutdown. ^{55}Fe is a hard-to-measure radionuclide…"

Ni 59
7.5 · 10^4 a
ε; β^+ ...
no γ; σ 77.7
$\sigma_{n,\alpha}$ 14; $\sigma_{n,p}$ 2
σ_{abs} 92

" **^{59}Ni** is considered an important radionuclide for waste disposal. This is a hard-to-measure radionuclide …"

Ca 41
1.03 · 10^5 a
ε; no γ
σ ~4
$\sigma_{n,\alpha}$ 0.18
$\sigma_{n,p}$ 0.007

" **^{41}Ca** can be of great importance when the safety of final disposal of decommissioning waste is assessed."

Cl 36
3.0 · 10^5 a
σ <10
$\sigma_{n,\alpha}$ 0.00059
$\sigma_{n,p}$ 0.046

" **^{36}Cl** …is important from the viewpoint of disposal, because of its long half-life, the solubility of chloride salts, low retardation in the geosphere and potential pathways to humans."

Zr 93
1.5 · 10^6 a
β^- 0.06...
m
σ <4

" **^{93}Zr** is also considered one of the critical radionuclides for long term disposal."

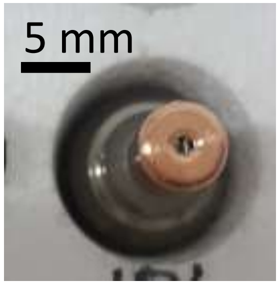
in 1998: only counting techniques considered for these radionuclides

→ since 1998 AMS technique became cheaper & more common

Samples used in the study

(Merchel et al., in review at J. Radioanalytical & Nuclear Chemistry)

- 2 fine and 2 coarse grained steel chips drilled or rasped from a nuclear reactor vessel
- ICP-MS: measurement: steel samples can be considered 100 % Fe
  → simpler transformation of measured $^{55}Fe/Fe$ AMS ratios to specific activities

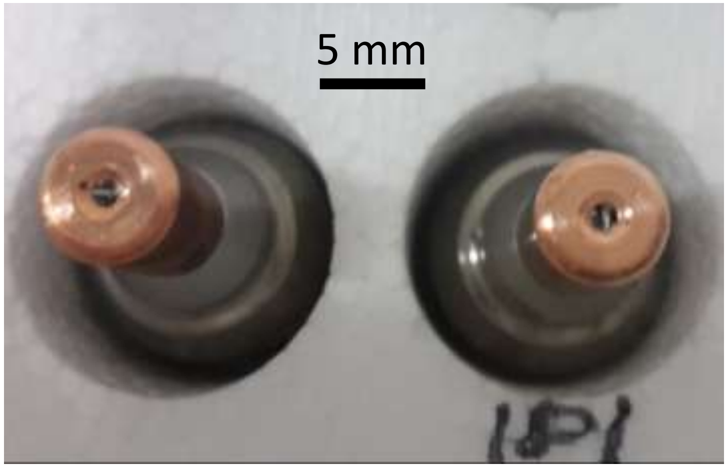


<u>alternative preparations for each sample:</u>

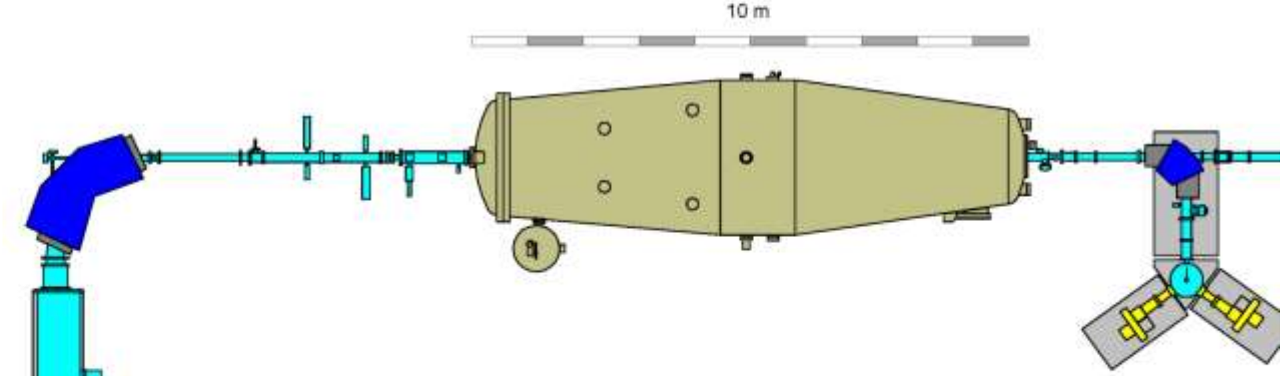
a. steel chips (mg amounts) directly hammered in 3 “targets”
b. radiochemical separation on bigger solid aliquots (~1.5g): hydroxide precipitations and extraction chromatography or liquid-liquid extraction
   → 96 % used for LSC analysis
   → 4 % for Fe_2O_3 AMS target

→ “5” results per sample

^{55}Fe "hard-to-measure"? – in steel it's an easy preparation for AMS !

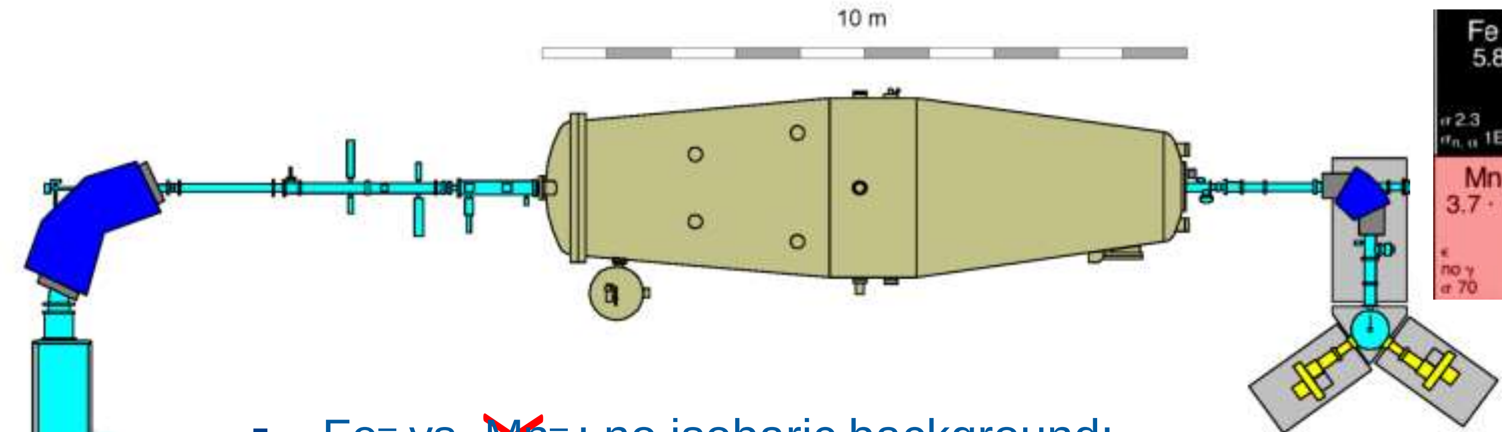


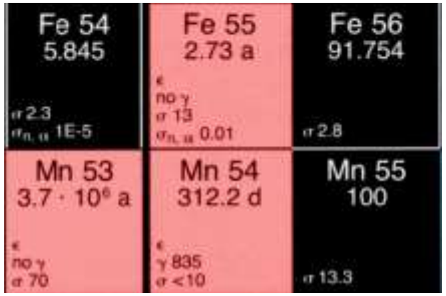
Introduction to DREsden AMS (DREAMS)



- negative ions (atomic or molecular) are extracted in Cs sputter ion source
- 1. mass separation of negative ions: Fe^{-}
- acceleration (voltage up to 6MV) & destruction of molecular background: stripping to positive charge states
- 2. mass separation of positive ions: Fe^{3+}
- measurement of ion currents (abundant isotopes) and detection of single ions in gas ionization detector (rare isotopes)
  → ratios $^{55}Fe/^{54}Fe$ and/or $^{55}Fe/^{56}Fe$

^{55}Fe “hard-to-measure”? – an easy measurement for AMS !

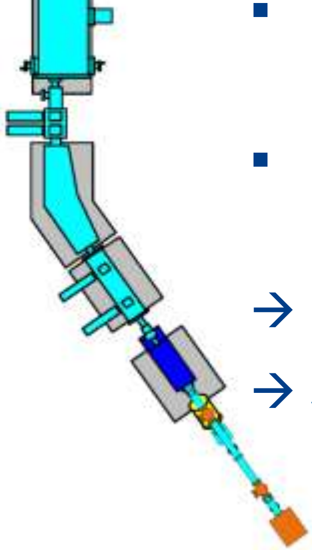


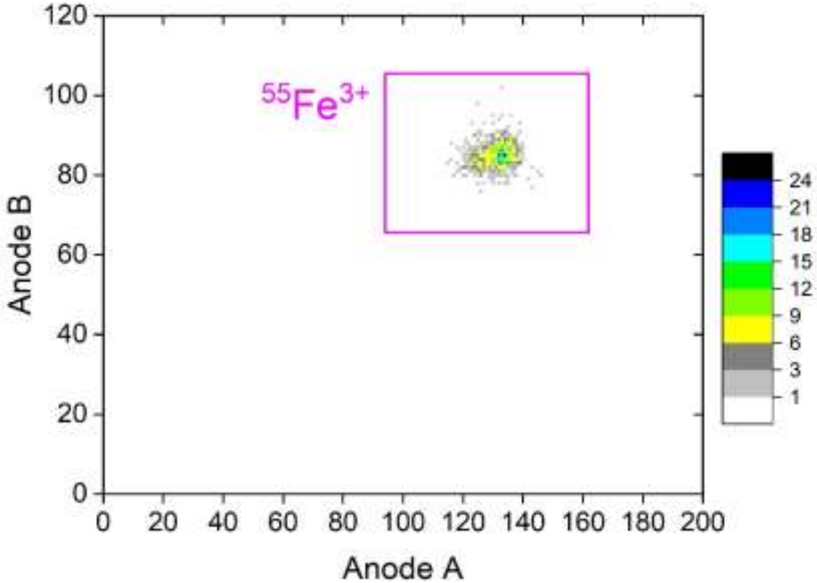


- Fe⁻ vs. ~~Mn~~⁻ : no isobaric background: ^{55}Mn does not form negative ions!
- only molecular isobars have to be suppressed (e.g. ^{54}FeH, ^{37}Cl^{18}O)

→ clean $^{55}Fe^{3+}$ spectrum in detector

→ simple setup of the machine





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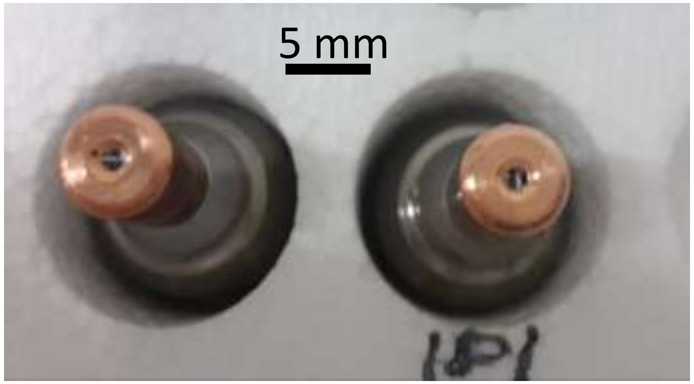
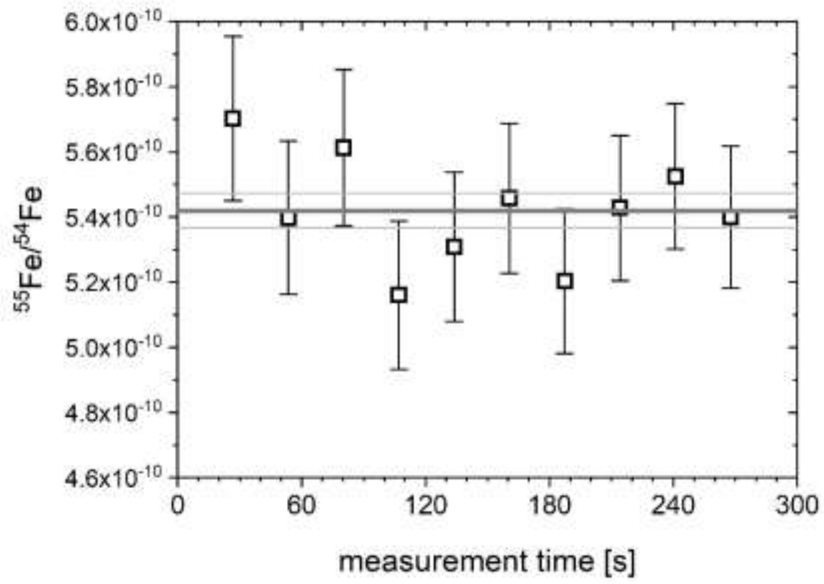
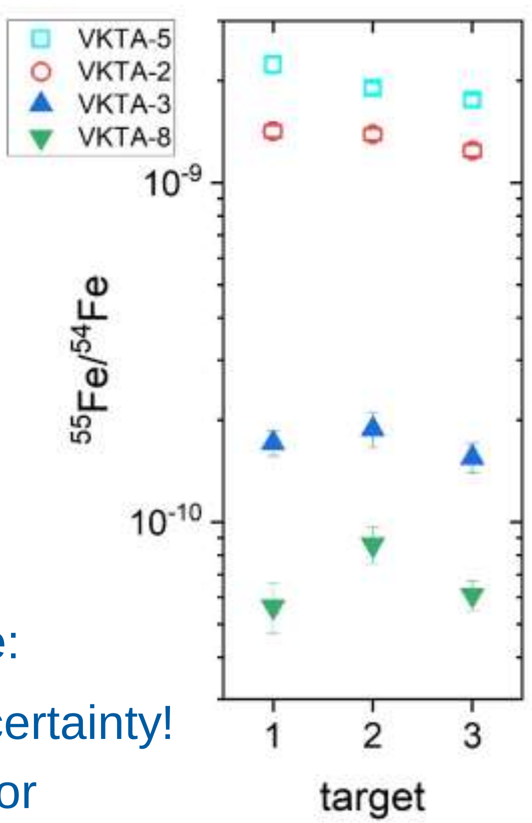
AMS-performance of steel samples



stability of a single measurement

→ overall uncertainty dominated by counting statistics!





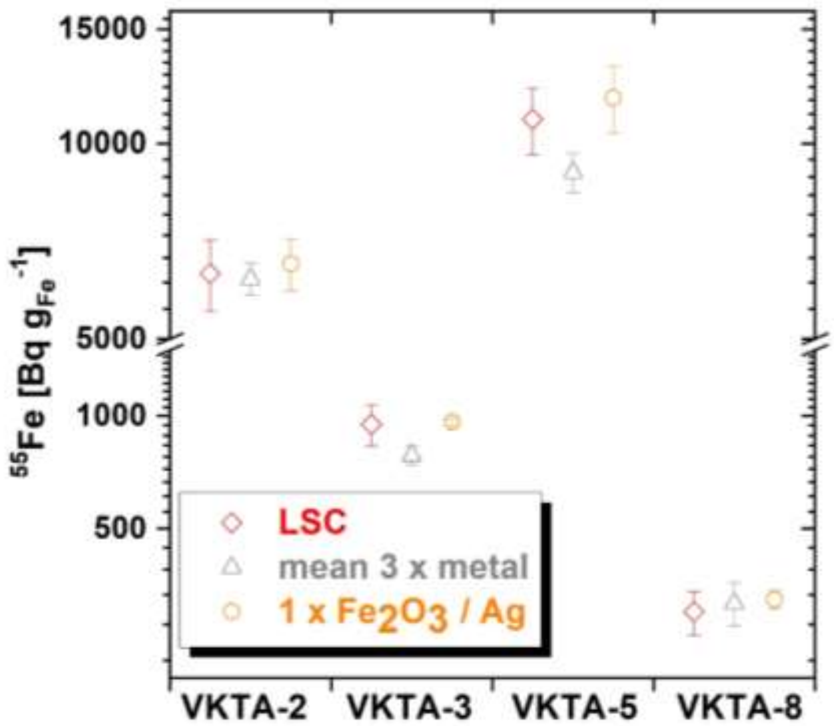
measurements of 3 targets for each steel sample:

→ scatter between targets dominates overall uncertainty!

➔ inhomogeneous steel flakes are problematic for high-precision measurements

AMS-LSC comparison

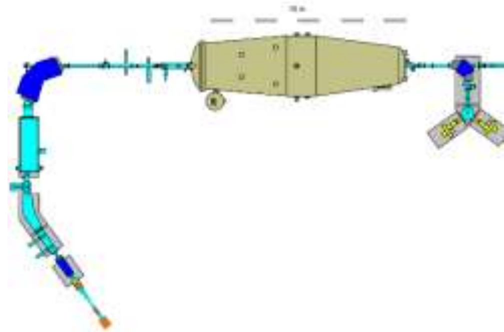
- linearity over > 2 orders of magnitude documented
- uncertainties of AMS competitive to LSC
- AMS background at single Bq / g_{Fe}



Merchel et al.,
J. Radioanalytical & Nuclear Chemistry,
in review

	AMS	LSC
time consumption for preparation (batch of 8 steel samples)	1 hour	20 hours over ca. 4 days
measurement time per sample	15 mins	150 mins
measurement preparation time (e.g. sample loading + tuning)	6 hours	<15 mins

Funding by Helmholtz Association and European Union



- HZDR ion beam centre (IBC) offers access to its experimental facilities free of charge
  → submit your project via HZDR “GATE”:
  scientific case + experimental plan
  evaluation by external board
  ➔ https://gate.hzdr.de/user/

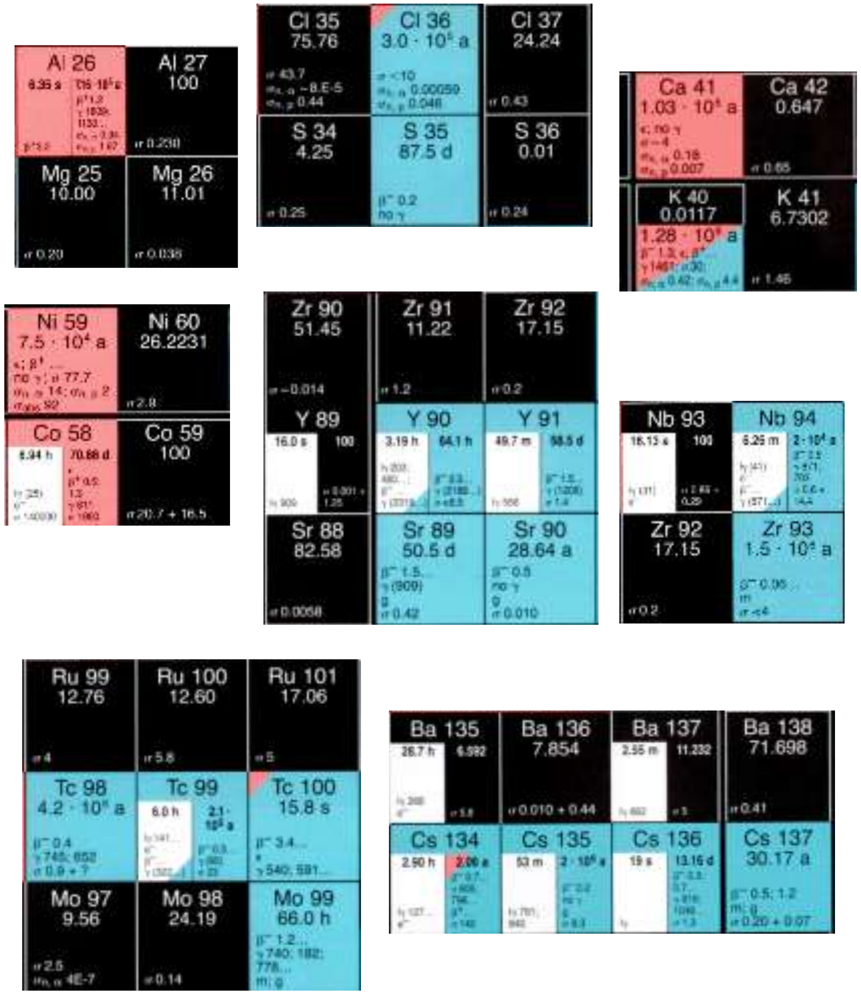
HELMHOLTZ
RESEARCH FOR GRAND CHALLENGES

- EU funds Trans-National-Access via HORIZON 2020 project RADIATE
  relevant for users working in EU member states (except Germany) and associated states
  → submit your project for evaluation by external advisory board
  → covers cost for beam time, travel & accommodation until 12/2022
  ➔ https://www.ionbeamcenters.eu/radiate/radiate-transnational-access/

RADIATE

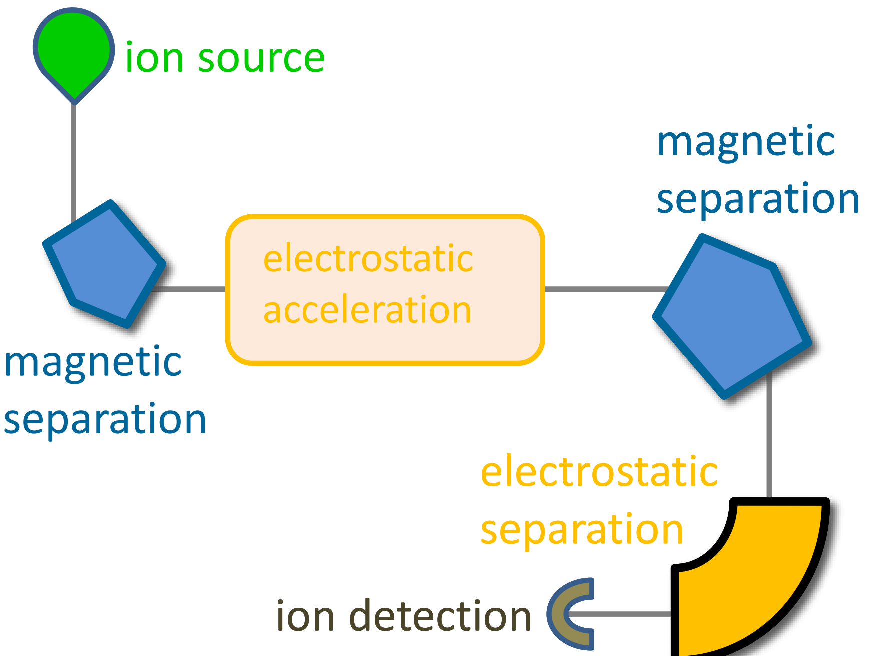
→ please contact j.lachner@hzdr.de

New AMS facility planned at HZDR: Start in 2023



Key features:

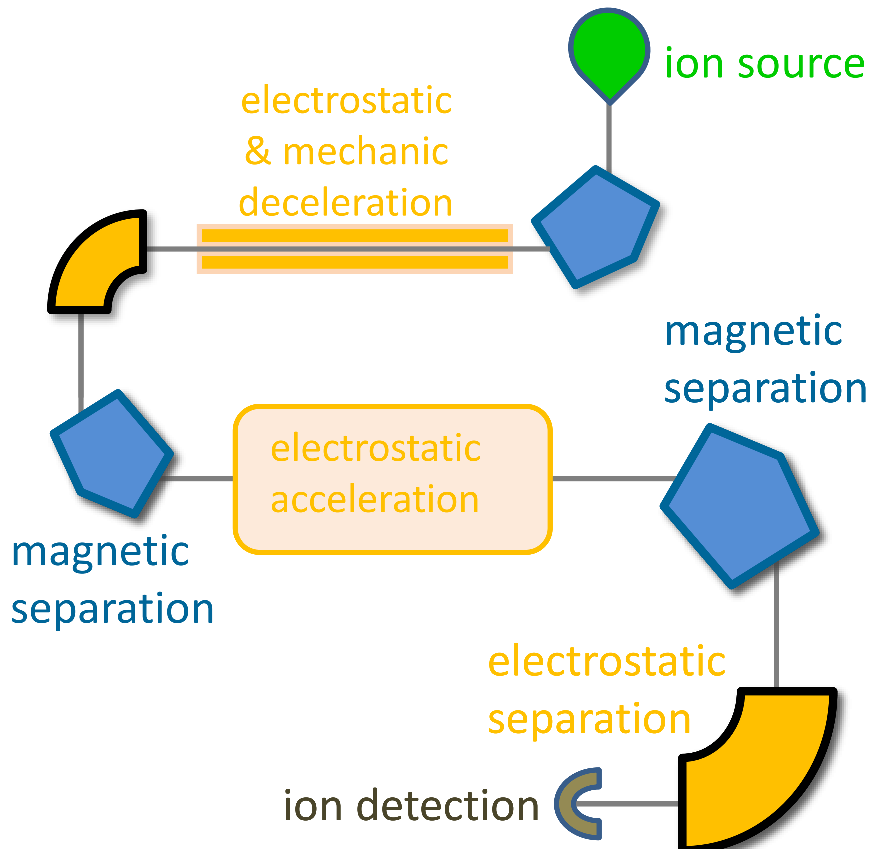
- smaller than DREAMS: accelerator max. 1MV
- dedicated AMS facility:
  easier access & faster sample turnaround
- focus on actinides and new AMS isotopes



Beyond present standards of AMS: ILIAMS – development of AMS towards analysis of ^{59}Ni, ^{93}Zr, etc.

- deceleration step of ions before injection into accelerator
- requires high-voltage platform on ion source potential
- slow ions have time for
  - physical interaction
  - chemical transformation

Collaboration with
M. Martschini, O. Marchhart,
A. Wieser, P. Steier, R. Golser



ILIAMS – development of AMS

$N_{surv} = N_0 \cdot e^{-\sigma\phi t}$;

σ: detachment cross section,

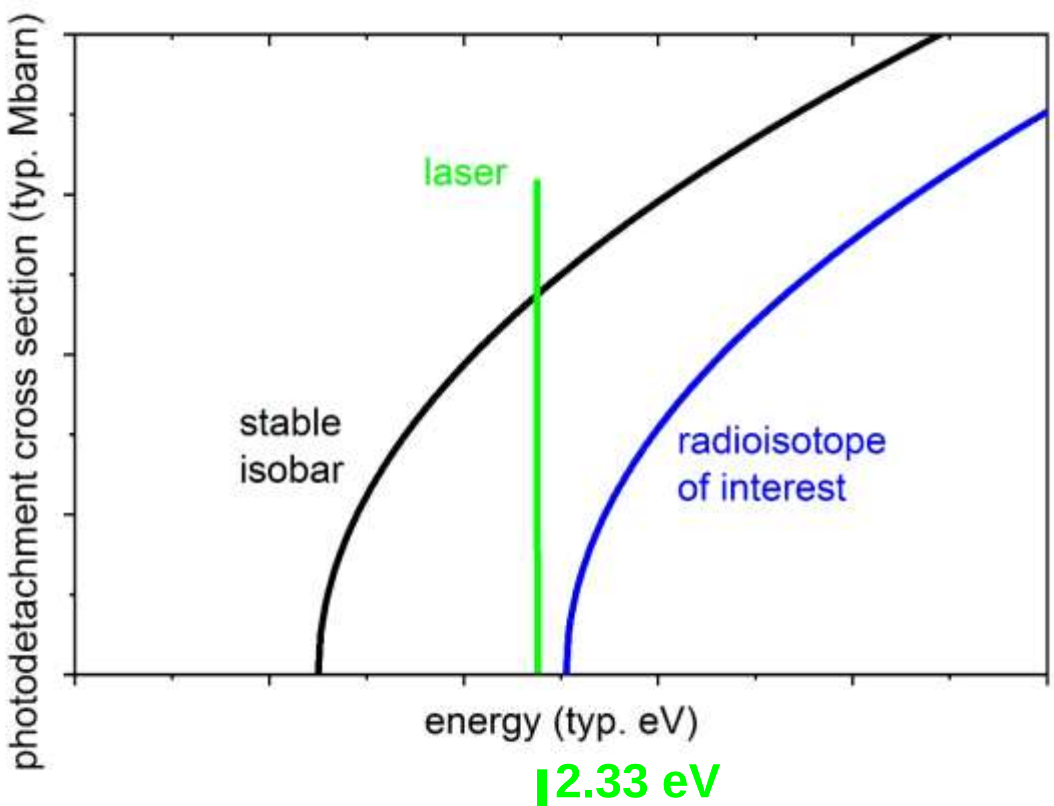
ϕ: photon flux,

t: interaction time

green laser: 18 W cw

532 nm: E_{ph}=2.33 eV

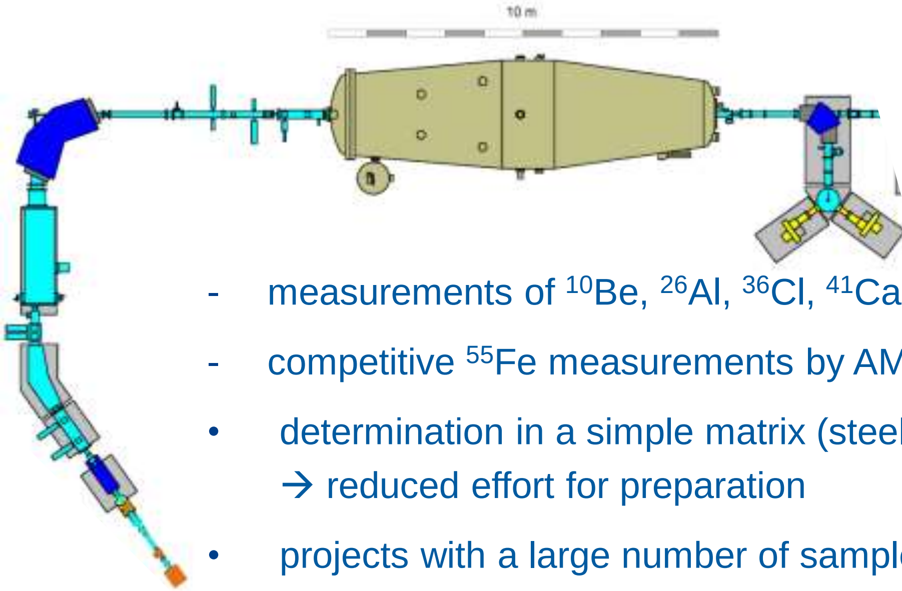
- ✓ S^- vs. Cl^-
- ✓ MgO^- vs. AlO^-
- ✓ BaF_2^- vs. CsF_2^-
- ✓ ZrF_3^- & YF_3^- vs. SrF_3^-



m=36	S^-: 2.0771029(10) eV	Cl^-: 3.612724(27) eV
m=43	MgO^-: 1.630(25) eV	AlO^-: 2.600(10) eV
m=90	ZrF_3^-,YF_3^-: < 2.3 eV	SrF_3^-: > 2.3 eV
m=173,175	BaF_2^-: < 2.3 eV	CsF_2^-: > 2.3 eV

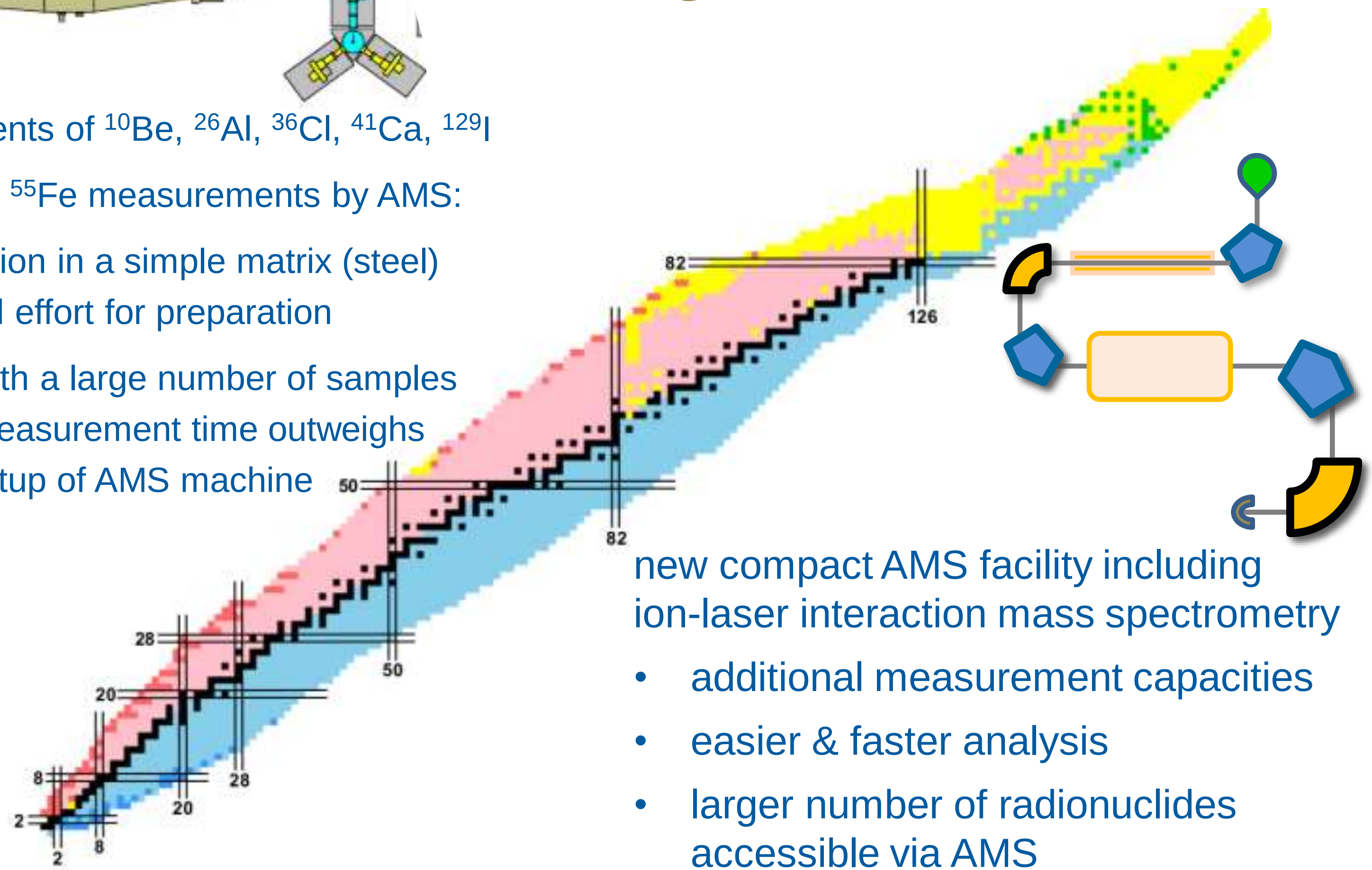
→ ^{36}Cl: Lachner et al., NIMB 2019, ^{26}Al: Lachner et al. IJMS 2021

AMS at HZDR: standard AMS radionuclides, ^{55}Fe and beyond



- measurements of ^{10}Be, ^{26}Al, ^{36}Cl, ^{41}Ca, ^{129}I
- competitive ^{55}Fe measurements by AMS:
  - determination in a simple matrix (steel)
    → reduced effort for preparation
  - projects with a large number of samples
    → short measurement time outweighs time for setup of AMS machine

DREAMS @ HZDR



new compact AMS facility including ion-laser interaction mass spectrometry

- additional measurement capacities
- easier & faster analysis
- larger number of radionuclides accessible via AMS

check for more details: www.hzdr.de/fwir