60Fe supernova-searches with AMS

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Russbach School
March 2014

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Outline

• Accelerator Mass Spectrometry (AMS)
  → Basics
  → AMS isotopes
  → Applications
• AMS Setups at MLL in Garching
• Recent example: Search for supernova 60Fe
  → in ferromanganese crusts
  → in microfossils
  → on the moon
• Summary and outlook
Accelerator Mass Spectrometry (AMS) is a high-sensitivity ion counting technique, primarily used for determination of isotopic ratios involving long-lived radionuclides.

- I will not be talking about $^{14}$C table top machines!
- Tandem-accelerator based systems → complete suppression of molecular background
- High energies (100-200 MeV) → nuclear physics particle identification techniques
- Sensitivity can reach down to isotopic ratios of $10^{-16}$
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- High energies (100-200 MeV) → nuclear physics particle identification techniques
- Sensitivity can reach down to isotopic ratios of $10^{-16}$
- Only few milligrams of sample material required (however: destructive!)
- Isotopic ratios measured → systematic errors (ion source efficiency, …) cancelled
- Challenge: Suppression of isobaric background, e.g. $^{60}$Ni, $^{60}$Fe or $^{53}$Cr, $^{53}$Mn, …
- AMS Facility used for all measurements in this study: Maier-Leibnitz-Laboratory Garching
AMS Basics

Sample (Element X)

Radioisotope $^nX$
Tiny fraction: $^nX/X = 10^{-12} \ldots 10^{-17}$
Sample (Element X)

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Ion beam → Tandem accelerator

$X^- \rightarrow X^{m+}$
Molecular background destroyed (gas or foil stripping)
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Molecular background destroyed (gas or foil stripping)

1.) Magnetic analyzer 
(typically 90 degree dipole magnet)

\[ E(v) = \frac{1}{2} q r B \cdot v \sim v \]

2.) Electrostatic analyzer (ESA)

\[ \frac{E}{q} = \frac{1}{2} \frac{r U}{d} \sim \text{const} \]

3.) Wien-Filter

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Faraday cup:
Number of stable X
Sample (Element X)

Radioisotope \(^n\text{X}\)

Tiny fraction: \(^n\text{X}/\text{X} = 10^{-12} \ldots 10^{-17}\)

Ion beam → Tandem accelerator

Molecular background destroyed (gas or foil stripping)

Filter elements (magnetic, electrostatic)

Faraday cup: Number of stable X

Detector (Particle Identification) e.g. TOF, E, E loss → number of \(^n\text{X}\)
AMS Basics

Sample (Element X)

Radioisotope $^nX$
Tiny fraction: $^nX/X = 10^{-12}...10^{-17}$

Problem: Transmission from Faraday cup to detector

→ Normally, AMS does **not** measure **absolute**!
→ compare number of counts to a standard sample
→ Give concentration **relative to standard sample**

Filter elements (magnetic, electrostatic)

Faraday cup: Number of stable X

Detector (Particle Identification) e.g. TOF, E, E loss
→ number of $^nX$
AMS Basics

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- Ion beam
- Tandem accelerator
- Molecular background destroyed (gas or foil stripping)

Filter elements (magnetic, electrostatic)

- Filter elements
- Faraday cup: Number of stable X

Typical sensitivity:

Current from ion source >1 µA ≈ 1E13 particles per s
Total transmission ≈ 10% (includes stripping yield)
Measuring time ≈ 1E4 s

→ checked particles X = 1E16
→ 1 count of \(^nX\) would yield \(\frac{nX}{X}=1E-16\)

This is a pretty hard limitation, because of limited beamtime

Detector (Particle Identification) e.g. TOF, E, E loss
→ number of \(^nX\)
AMS requirements

Can I measure my favourite isotope with AMS?
AMS requirements

Half-life:
\[ T_{1/2} > 100 \text{ Ma} \rightarrow \text{primordial background} \]
\[ T_{1/2} < \text{weeks} \rightarrow \text{better use decay counting} \]

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- Does your isotope have a stable isobar?
  - e.g. $^{53}\text{Mn}$ and $^{53}\text{Cr}$ → setup needs isobar suppression
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Production of negative ion beam possible?
(Required for tandem accelerators)
Higher current → better sensitivity
AMS requirements

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- **Can I measure my favourite isotope with AMS?**

- **Production of negative ion beam possible?**
  - (Required for tandem accelerators)
  - Higher current $\rightarrow$ better sensitivity

- **Standard material available?**
  - Since AMS measures relative, standard necessary to obtain concentrations
AMS isotopes

Fig. 4 from D. Fink, NIM B 268, 2010
modified/updated from W. Kutschera 1981
AMS is multidisciplinary

- Environmental and Geo-science
  - atmosphere, oceanography, glacier, **climate**, groundwater, erosion, dating paleooceanography, …

- Material science
  - fusion research, active waist management

- Human metabolism and medical application
  - **dosimetry**, pharmacology, …

- Chemistry
  - tracing molecules and elements, …

- Extraterrestrial
  - meteorites (e.g. Lunar and Martian origin)
  - **interplanetary dust, SN**, related cross sections, …

- Physics
  - **nuclear astrophysics**, super- asymmetric fission, …
Maier-Leibnitz Laboratory (MLL) Garching
Isobar suppression: GAMS

Challenge: Isobar separation of $^{60}$Ni → use of the Gas-filled-Analyzing-Magnet-System (GAMS)
Ionization chamber

9 „independent“ signals

e5
e4
e3
e2l
e2r
e1l
e1r

Frisch grid: et
y-angle: dt

position: p
x-angle: dp
Ionization chamber
60Fe standard sample $^{60}\text{Fe}/^{56}\text{Fe} \approx 1\times 10^{-12}$
Real sample $^{60}\text{Fe}/\text{Fe}=?$
Real sample $^{60}\text{Fe}/\text{Fe} =$?
Real sample $^{60}\text{Fe}/\text{Fe}=$?
## AMS measurements with GAMS

<table>
<thead>
<tr>
<th>isotope</th>
<th>application</th>
<th>ion in ion out</th>
<th>isobar</th>
<th>isobar Supp.</th>
<th>sensitivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{26}$Al</td>
<td>astroph. x-section manganese crusts</td>
<td>$^{26}\text{Al}$- $^{26}\text{Al}^{7+}$</td>
<td>$^{26}\text{Mg}$</td>
<td>$10^9$</td>
<td>few $10^{-15}$</td>
</tr>
<tr>
<td>$^{36}$Cl</td>
<td>groundwater, meteorites</td>
<td>$^{36}\text{Cl}$ - $^{36}\text{Cl}^{19+}$</td>
<td>$^{36}\text{S}$</td>
<td>$10^9$ - $10^{10}$</td>
<td>$&lt; 10^{-15}$</td>
</tr>
<tr>
<td>$^{41}$Ca</td>
<td>Hiroshima teeth, meteorites</td>
<td>$^{41}\text{CaH}_3$, $^{41}\text{CaF}_3$ - $^{41}\text{Ca}^{3+}$</td>
<td>$^{41}\text{K}$</td>
<td>$10^9$ - $10^{10}$</td>
<td>$&lt; 10^{-15}$ $10^{-14}$ CaF</td>
</tr>
<tr>
<td>$^{44}$Ti</td>
<td>astroph. x-section</td>
<td>$^{44}\text{TiO}$ - $^{44}\text{Ti}^{9+}$</td>
<td>$^{44}\text{Ca}$</td>
<td>$10^9$ - $10^{10}$</td>
<td>few $10^{-15}$</td>
</tr>
<tr>
<td>$^{52}$Mn</td>
<td>crusts, in-situ, meteorites, sediments, moon rocks</td>
<td>$^{52}\text{Mn}^{17+}$</td>
<td>$^{52}\text{MnF}$ - $^{52}\text{Mn}^{11+}$</td>
<td>$^{52}\text{Cr}$</td>
<td>$10^9$</td>
</tr>
<tr>
<td>$^{59}$Ni</td>
<td>nuclear waste, meteorites, astroph. x-section</td>
<td>$^{59}\text{Ni}$ - $^{59}\text{Ni}^{12+}$</td>
<td>$^{59}\text{Co}$</td>
<td>$10^9$</td>
<td>few $10^{-14}$</td>
</tr>
<tr>
<td>$^{60}$Fe</td>
<td>crusts, meteorites, moon rock, nuclear waste</td>
<td>$^{60}\text{FeO}$ - $^{60}\text{Fe}^{11+}$</td>
<td>$^{60}\text{Ni}$</td>
<td>$&gt;10^{11}$</td>
<td>few $10^{-16}$</td>
</tr>
<tr>
<td>$^{63}$Ni</td>
<td>Hiroshima copper, neutron dosimetry astroph. x-section</td>
<td>$^{63}\text{Ni}$ - $^{63}\text{Ni}^{12+}$</td>
<td>$^{63}\text{Cu}$</td>
<td>$10^3$ - $10^9$</td>
<td>few $10^{-14}$</td>
</tr>
<tr>
<td>$^{72, 93}$Se, $^{107}$Pd</td>
<td>astroph. x-section, half-life, nuclear waste</td>
<td>various</td>
<td>$^{72}\text{Br}$, $^{93}\text{Nb}$, $^{107}\text{Ag}$</td>
<td>$10^3$</td>
<td>few $10^{-12}$ ? $10^{-16}$ ?</td>
</tr>
</tbody>
</table>
Chosing an isotope

However, I can only talk about ONE of those isotopes today:
Why $^{60}\text{Fe}$?

![Graph showing the abundance of $^{60}\text{Fe}$ in different nuclear charge regions.

- CR spallation on air
- CR spallation on extraterrestrial material
- Nuclear fission products

- Solar abundances
- Fe 60
  - $1.5 \times 10^6$ a
  - $\beta^- 0.1$}
Why $^{60}\text{Fe}$?

→ Unique situation: 1) Isobar $^{60}\text{Ni}$ has $\Delta Z = 2$ (good for AMS!)
   2) Expected background in terrestrial samples $^{60}\text{Fe}/\text{Fe} < 10^{-16}$
   3) Half-life on the order of Myr (interesting SN ages)
Motivation

Massive star forming $^{60}$Fe

Betelgeuse (Hubble)
Motivation

Supernova explosion

$^{60}\text{Fe}$

Betelgeuse (Hubble)

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Interstellar medium near Earth:
Local Bubble

Massive star forming $^{60}$Fe

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Interstellar medium near Earth: Local Bubble

Solar system can „pick up“ supernova debris

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Ferromanganese Crust

Marine Sediment

Lunar samples

Massive star forming $^{60}$Fe
Earlier measurement: Ferromanganese Crust

Ferromanganese crust from equatorial pacific (9°18´N, 146°03´W), depth 4830 m

- Slow growing (few mm per Ma)
- Can be dated by 10Be measurements
Earlier measurement: Ferromanganese Crust

Ferromanganese crust from equatorial pacific
(9°18´N, 146°03´W), depth 4830 m
Earlier measurement: Ferromanganese Crust

- Indication for a supernova signal already found using the GAMS setup
- Possible SN event 2-3 Ma ago at a distance ~40 pc
- Goal: Confirmation of this finding in slow growing sediment

Ferromanganese crust from equatorial pacific (9°18´N, 146°03´W), depth 4830 m

Knie et. al., PRL 93, 171103 (2004)
Earlier measurement: Ferromanganese Crust

Ferromanganese crust from equatorial pacific (9°18´N, 146°03´W), depth 4830 m

Knie et. al., PRL 93, 171103 (2004)
Search in Pacific Ocean sediment

Search for $^{60}$Fe in ocean sediment:

- Samples from two drill cores from ODP (Ocean Drilling Program) were obtained, 8 kg of material total – leg 138 – Cores 848 + 851
- Goal: measure depth profile of $^{60}$Fe/Fe with resolution $\sim$30,000 years in the age range 1.8 – 3.8 Myr
- Dating available from magnetic field reversal (among others), however, independent dating currently underway using 10Be and 26Al at DREAMS in Dresden
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Problems:
- Which Fe-bearing minerals could contain the $^{60}$Fe signal?
- How to prevent dilution of signal?

Primary
Transported in by wind/water
Large grains ($\mu$m)
$^{60}$Fe only on surface

Secondary
Formed in situ
Small grains (20-200 nm)
e.g. magnetite crystals ($\text{Fe}_3\text{O}_4$)
Magnetotactic bacteria

Intracellular formation of biogenic magnetite crystals:

- Magnetotactic bacteria live in sediment just below the surface-water interface

- Bacteria build up chains of magnetite grains (20-80 nm) for orientation in earth's magnetic field (magnetotaxis) → secondary!

- In $^{60}$Fe-enriched ocean water, bacteria are forced to build magnetosomes with $^{60}$Fe-rich iron

- Magnetic signature can be preserved over geologically significant timescales if the magnetosome chains survive sedimentation.

- Magnetic signature resembles non-interacting single domain (SD) particles → characterization using magnetic measurements is possible because of their unique properties

Sediment sample – 150 Myr after sedimentation → chain structure still visible
TEM on Sediment

Transmission electron microscopy (TEM):

- After magnetic extraction on one of our sediment samples
- High abundance of small magnetite grains as chain fragments and clusters
Interpretation:

- Is this magnetite? → zoom in → diffraction analysis OR perform EDX, both say: YES

- It it biogenic? Both shape and size say YES

- How much is it? → roughly 10% (mass) of the magnetic extract
  → fits very well with estimate from preliminary ARM/IRM magnetic measurements

TEM on Sediment
How to get small grained magnetite out to make AMS samples?

→ Citrate Bicarbonate Dithionite technique (CBD)
  – very mild leaching (dissolves only < 200 nm)
  – 30 g of sediment yield about 5 mg of Fe2O3 AMS sample
  – about 1 week of chemistry needed (not shown here, just to mention...)
AMS results core 848

Sediment core 848 - 60Fe/Fe results

Blank level (1-σ): $^{60}$Fe/Fe = 1.7e-16
From chemistry blank (0 counts)
AMS results core 848

Sediment core 848 – 3 samples grouped each – 8 counts total

---

Blank level (1-σ): $^{60}\text{Fe}/\text{Fe} = 1.7 \times 10^{-16}$
From chemistry blank (0 counts)
Core 851 preliminary results

- Data analysis still ongoing, better statistics will be available soon
- The signal is not yet significant enough → more beamtimes necessary
- 12 counts total detected so far
Preliminary results from sediment cores

CORE 848

- Core sediment used up except for aliquots
- 90% of AMS samples already measured
- 8 counts of 60Fe detected in the range 1.9 – 2.7 Ma
- Average concentration over 1.9 – 2.6 Ma:
  \[ \frac{^{60}\text{Fe}}{\text{Fe}} \approx 1 \times 10^{-15} \]
- Rough estimation of total number of incident 60Fe atoms can be made
  \[ \Phi_{\text{LIF}} \approx \text{few } 10^7 \quad ^{60}\text{Fe cm}^{-2} \]
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  → local interstellar fluence
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CORE 851
- Only ~30% of core measured yet
- 13 counts of $^{60}$Fe detected, among those 12 in the range 1.7-2.5 Ma
- Average concentration in that range only
  \[ 2-3 \times 10^{-16} \]
- This is very low → better blank level needed and of course, more counts $^{60}$Fe
- Idea: try magnetic extraction instead of chemical to reduce dilution → currently being set up
## Preliminary results from sediment cores

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<tr>
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<th>CORE 851</th>
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<td>7 counts of $^{60}$Fe detected in the range 1.9 – 2.7 Ma</td>
<td>Average concentration in that range only 2-3 x $10^{-16}$</td>
</tr>
<tr>
<td>Average concentration over 1.9 – 2.6 Ma: $^{60}$Fe/Fe ~ $1x10^{-15}$</td>
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<td>→ local interstellar fluence $\Phi_{\text{LIF}}$ ~ few $10^7$ $^{60}$Fe cm$^{-2}$</td>
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Most important result so far: **$^{60}$Fe input** seems to have been **rather long (> 500 kyr)**. This was previously unknown from crust measurements because of low time resolution.
$^{60}\text{Fe}$ (and $^{53}\text{Mn}$) in lunar samples
Apollo Landing Sites

- 60Fe and 53Mn Samples from Apollo 12, 15, and 16 obtained

Lunar Reconnaissance Orbiter Camera
Arizona State University, November 2011
On the moon...

- Net sedimentation rates are small: U60Fe~100%
- Ni concentrations are in general low: low in-situ production of 60Fe

- Gardening of the lunar surface: ~2-3 cm reworking depth in 10Myr
- Hard to reach

---

Sample history and production of $^{60}$Fe

Potential origins:
- Galactic Cosmic Rays
- Solar Cosmic Rays
- Deposition of SN debris.

$P_{^{60}Fe}-GCR = (0.88 \pm 0.44)$ dpm/kg Ni


$P_{^{60}Fe}-SCR = 0.07–0.45$ dpm/kg Ni on the surface

Cook et al, LPSC XXXIX (2009)
Plot shows both radioisotopes, 53Mn and 60Fe divided by mass of their respective most likely source element under irradiation from cosmic rays.

Enhancement in both isotopes for some samples visible.
60Fe and 53Mn measurements

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- Enhancement in both isotopes for some samples visible.

Expected cosmogenic contribution

- 1-2: Apollo 12
- 3-4: Apollo 16 shaded
- 5-7: Apollo 15
- 8-11: Apollo 16
- 12-18: Meteorites
60Fe and 53Mn measurements

- Excess in 60Fe in 5 samples interpreted as supernova input

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Excess in 60Fe in 5 samples interpreted as supernova input

Ingetrate to obtain Local interstellar fluence:

\[ \Phi_{60\text{Fe, LIS}} \approx 3.5 \times 10^7 \text{ at} \text{cm}^{-2} \]
Summary

- AMS is an ultrasensitive technique for isotope ratio measurement
- Sensitivities reaching down to and below 1E-16
- Applications in nuclear astrophysics
- Setup at MLL in Garching with 2 AMS beamlines
- GAMS beamline for isobar suppression (intermediate A)
- New results: 60Fe signature found in sediment (~20 counts so far but: more statistics required → further beamtimes → favors long input time (~500 kyr)
- Lunar samples scanned for 53Mn and 60Fe
- Enhancement of 60Fe found in 5 samples → SN input
  → $\phi_{\text{LIS}} \approx 3.5 \times 10^7$ at/cm$^2$ (not decay-corrected)
Thank you for your attention
Fe content of drill core material ~1-2%

$^{60}$Fe expected in small iron oxide (e.g. magnetite) grains: size $<< 1 \mu m$ in the form of bacteria fossils and others

Primary iron oxide (brown) would dilute the signal contained in secondary minerals (black)

Chemical extraction of small grained iron oxides using Citrate-bicarbonate-dithionite (CBD) extraction method: Dissolves mainly small grained material

$\rightarrow$ Dithionite: Strong reducing agent
   (Fe $III^+$ $\rightarrow$ Fe $II^+$)
$\rightarrow$ Citrate: extracts and chelates Fe $II^+$
$\rightarrow$ Sodium-Bicarbonate: pH buffer
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\[\text{Dissolved by CBD}\]

\[\text{Not dissolved}\]
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- $^{60}$Fe expected in small iron oxide (e.g. magnetite) grains: size $< 1 \mu$m in the form of bacteria fossils and others

- Primary iron oxide (brown) would dilute the signal contained in secondary minerals (black)

- Chemical extraction of small grained iron oxides using Citrate-bicarbonate-dithionite (CBD) extraction method: Dissolves mainly small grained material
  
  - Dithionite: Strong reducing agent ($\text{Fe}^{\text{III}}+ \rightarrow \text{Fe}^{\text{II}}+$)
  
  - Citrate: extracts and chelates Fe II+
  
  - Sodium-Bicarbonate: pH buffer

- ~ 5 mg Fe$_2$O$_3$ AMS sample

- Not dissolved

- 200 nm
New 60Fe measurements

- New 60Fe measurements show 60Fe counts in 7 samples
- To distinguish from cosmic ray production look at 60Fe/kg(Ni)
  - → 3 samples show significantly higher values than highest cosmogenic production observed in meteorites
  - → identification as SN input possible
Cosmic ray spallation:

- $^{60}$Fe production in cosmic ray spallation mostly on Ni targets
- On Earth, atmosphere prevents large build-up - background $^{60}$Fe/Fe < $10^{-16}$
- In meteorites and lunar samples, this is not given
Production in stars:

- 2 Neutron captures (s-process) on $^{58}$Fe
- Shell He burning in massive stars ($M > 40 \, M_{\odot}$)
- Shell C burning in massive stars ($M < 40 \, M_{\odot}$)
- Explosive synthesis in SN when shockwave passes through shells $\rightarrow$ small contribution

Magnetite in sediment

“Extracellular magnetite”
Secondary magnetite precipitated from redox reactions, either *inorganically*, or by mediation of *dissimilatory metal reducing bacteria* (DIRB)

“Biogenic magnetite”
Magnetosomes and magnetosome chains produced by *magnetotactic bacteria* (MB)
Do we have biogenic magnetite in our sediment?

Transmission electron microscopy (TEM):

- Setup at TUM Chemistry (Marianne Hanzlik)
- First try: TEM on untreated sediment
  → extremely difficult and time consuming
- Results show at least some biogenically-looking magnetite particles (magnetite identified by diffraction analysis)
Transmission electron microscopy (TEM):

- Second try: pre-treatment of sediment with AcOH (20%) for 1 hour → reduction of sample mass by 80% → calcite matrix dissolved
- Still extremely time-consuming and difficult
TEM on Sediment

Transmission electron microscopy (TEM):

- Third try: Insert magnetic finger into sample from try #2
- Much easier to find magnetite formations: chains and clusters can be seen
- HOWEVER: Are these originally present in the sediment? Or just formed during magnetic extraction? → impossible to interpret
Future Plans for Sediment samples

- Attempt to obtain more sediment of core 848 in SN region
  → Currently being discussed, samples may be available

- Try alternative extraction method:
  → Dissolve calcite matrix with AcOH (20%)
  → Perform magnetic extraction
  → Optionally then CBD extraction
  → AMS

The magnetic extraction is currently being set up in Garching as a Bachelor's Thesis.
53Mn in lunar samples

Excess of $^{53}$Mn?

? Mixing of the regolith
? Addition of irradiated material
? Deposition of SN debris
? ...

AS12-49-7286
Transmission electron microscopy (TEM):

- Fourth try: Perform magnetic extraction directly on sediment

- VIDEO
FORC measurements

Use an Alternate Gradient Field Magnetometer (in Bremen) to record a set of 450 first-order reversal curves (FORCs), 4-5 times → total time ~1 day per sample
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment:

\[
\text{Fe}_3\text{O}_4 = \text{Fe}(\text{II}+) + \text{Fe}(\text{III}+)
\]

1h, 40 °C

CBD extraction

\[
\text{Fe}(\text{III}+) \rightarrow \text{Fe}(\text{II}+)
\]

Filtration: remove undissolved sediment
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment: \( \text{Fe}_3\text{O}_4 = \text{Fe}(\text{II}+) + \text{Fe}(\text{III}+) \)

CBD extraction \( \text{Fe}(\text{III}+) \rightarrow \text{Fe}(\text{II}+) \)

Filtration: remove undissolved sediment

\[ \text{Fe}(\text{II}+) \text{ Chelate In 200ml H}_2\text{O} \]

1h, 40 °C

Evaporation

300°C → Citrate decomposes
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment: $\text{Fe}_3\text{O}_4 = \text{Fe}^{(II+)} + \text{Fe}^{(III+)}$

1h, 40 °C

CBD extraction
$\text{Fe}^{(III+)} \rightarrow \text{Fe}^{(II+)}$

Filtration: remove undissolved sediment

$300^\circ\text{C} \rightarrow \text{Citrate decomposes}$

$\text{Fe}^{(II+)} \rightarrow \text{Fe}^{(III+)}$

+ $\text{HNO}_3$

Fe$^{(II+)}$ → Fe$^{(III+)}$

+ $\text{HNO}_3$

Fe$^{(II+)}$ → Fe$^{(III+)}$

+ $\text{HCl}$ → Fe$^{(III+)}$

Extracted + centrifuge

Fe$^{(III+)}$ in HCl

evaporation
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment: \( \text{Fe}_3\text{O}_4 = \text{Fe}(\text{II}^+) + \text{Fe}(\text{III}^+) \)

1h, 40 °C

CBD extraction
\( \text{Fe}(\text{III}^+) \rightarrow \text{Fe}(\text{II}^+) \)

Filtration: remove undissolved sediment

Fe(II+) Chelate
In 200ml H\(_2\)O

evaporation

Evaporation
Until dry
→ white residue

300°C → Citrate decomposes

+\( \text{HNO}_3 \)
\( \text{Fe}(\text{II}^+) \rightarrow \text{Fe}(\text{III}^+) \)

+\( \text{HCl} \) → Fe(III+) Extracted + centrifuge

Fe(III+) in HCl
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment: \( \text{Fe}_3\text{O}_4 = \text{Fe}(\text{II}+) + \text{Fe}(\text{III}+) \)

1h, 40 °C

CBD extraction

\( \text{Fe}(\text{III}+) \rightarrow \text{Fe}(\text{II}+) \)

Filtration: remove undissolved sediment

In 200ml \( \text{H}_2\text{O} \)

Evaporation

300°C → Citrate decomposes

\( +\text{HNO}_3 \)

\( \text{Fe}(\text{II}+) \rightarrow \text{Fe}(\text{III}+) \)

Evaporation Until dry → white residue

20 ml \( \text{HCl} \)

\( \text{Fe}(\text{III}+) \rightarrow \text{Fe}(\text{III}+) \)

Extracted + centrifuge

\( \text{Fe}(\text{III}+) \) in \( \text{HCl} \)
Chemical isolation of iron

Procedure for ~30g sediment

Magnetosomes in Sediment: $\text{Fe}_3\text{O}_4 = \text{Fe(II+)} + \text{Fe(III+)}$

Filtration: remove undissolved sediment

1h, 40 °C

CBD extraction
$\text{Fe(III+)} \rightarrow \text{Fe(II+)}$

Fe(II+) Chelate in 200ml $\text{H}_2\text{O}$

Evaporation

300°C → Citrate decomposes

$\text{Fe(II+)} \rightarrow \text{Fe(III+)}$

$+\text{HNO}_3 \rightarrow \text{Fe(III+)}$ Extracted + centrifuge

Evaporation

20 ml Hcl

Fe(III+) in $\text{HCl}$

Precipitate Fe as hydroxide with $\text{NH}_3$ (aq)

→ centrifuge
Chemical isolation of iron

Precipitate Fe as hydroxide with \( \text{NH}_3(\text{aq}) \)

\( \rightarrow \) centrifuge

Ion Exchange Column
Chemical isolation of iron

Precipitate Fe as hydroxide with $\text{NH}_3(\text{aq})$
→ centrifuge

Ion Exchange Column

Precipitate again, Wash with $\text{H}_2\text{O}$
Dry

≈ 3-5 mg $\text{Fe}_2\text{O}_3$
Crystal
Chemical isolation of iron

1. Precipitate Fe as hydroxide with \( \text{NH}_3(\text{aq}) \)
   → centrifuge

2. Ion Exchange Column

3. Precipitate again, Wash with H\(_2\)O
   Dry

4. ~ 3-5 mg \( \text{Fe}_2\text{O}_3 \)
   Crystal

5. Mix 50/50
   With Ag powder
   → Hammer into
   Ag AMS sample
   Holder → ready

6. Lehrstuhl E12
Hysteresis measurements

B (H) – External mag. Field [mT]
M – Magnetization

$M_s$ – saturation magnetization
$M_r$ ($M_{rs}$) – remanence magnetization

$H_c$ – Coercivity

$H_{cr}$ – Coercivity of remanence

$\chi_{lf}$ – low-field susceptibility

IRM – Isothermal remnant mag.

(a) Hysteresis loop of ferromagnetic material
(b) Difference between descending and ascending curve of (a), giving another measure for $H_{cr}$

Tauxe, L., Essentials of Paleomagnetism, University of California Press, 2010
Day plot:

- Domain-state sensitive
- Generated from bulk mag. Properties
- Problem: Non-unique interpretations
- Hints to high abundance of magnetofossils in our sediment, BUT:
  - For quantitative analysis, this is not enough!

Legend:

- SD – single domain
- MD – multi domain
- SP – super paramagnetic
- PSD – pseudo SD
- M – magnetite
- TM – Titanomagnetite
AMS sample coercivity distribution

Untreated sediment:
- \( \approx 2\% \) total Fe
- 60 ppm SD iron
- \( \approx 3\% \) Fe in mag. minerals

CBD extraction

Sample for AMS:
- 60% from magnetic minerals
- 40% from non-mag. minerals
- < 6% from primary mag. minerals
- >27% from mag. Bacteria

Magnetofossils !!!
Summary magnetic characterization core 848

Untreated sediment:
- \( \approx 2\% \) total Fe
- 60 ppm SD iron
- \( \approx 3\% \) Fe in mag. minerals

CBD extraction

Sample for AMS:
- 60\% from magnetic minerals
- 40\% from non-mag. minerals
- \(< 6\% \) from primary mag. minerals
- >27\% from mag. Bacteria

Assuming \(^{60}\text{Fe}\) only in secondary magnetic minerals (lower limit)
\(^{60}\text{Fe}/\text{Fe}\) is enhanced by a factor of \( \approx 250 \) relative to complete dissolution of Fe.

Dilution compared to original \(^{60}\text{Fe}\) concentration in secondary minerals \(< 1.9\)

\( \rightarrow \) core 848 suited for CBD extraction, assuming roughly constant composition across the core
FORC measurements

Use an Alternate Gradient Field Magnetometer (in Bremen) to record a set of 450 first-order reversal curves (FORCs), 4-5 times → total time ~1 day per sample

Mathematical transformation: \( H - M \rightarrow H_u - H_c \)
- \( H_u \): bias field (from other particles)
- \( H_c \): coercivity (mag. hardness)

Feature of FORC diagram of SD particles:
- Central ridge: Delta function along \( H_u = 0 \)
- to identify non-interacting SD particles, e.g. magnetofossils

Lake sample containing magnetofossils

\[ \rho(H_u, H_c) \]
\[ [10^{-4} \text{ Am}^2/\text{T}^2] \]
FORC measurements

Use an Alternate Gradient Field Magnetometer (in Bremen) to record a set of 450 first-order reversal curves (FORCs), 4-5 times → total time ~1 day per sample

Mathematical transformation: $H - M \rightarrow H_u - H$

where $H_u$ : bias field (from other particles)

$H_c$ : coercivity (mag. hardness)

Feature of FORC diagram of SD particles:
Central ridge: Delta function along $H_u = 0$
to identify non-interacting SD particles, e.g. magnetofossils

Integrate and fit coercivity distribution:
For the lake sample – 3 components
(1) Extracellular (low coercivity)
(2) Biogenic soft
(3) Biogenic hard
FORC analysis of sediment

Sediment 848 – 3.2-3.3 Myr untreated

Sediment 848 – 3.2-3.3 Myr after CBD extraction

Central ridge almost disappears (~5% left) after 1 CBD extraction

BUT: What's the magnetic signature of the AMS sample?
Central ridge almost disappears (~5% left) after 1 CBD extraction

BUT: What's the magnetic signature of the AMS sample?
FORC analysis of sediment

Sediment 848 – 3.2-3.3 Myr
untreated

Sediment 848 – 3.2-3.3 Myr
After CBD extraction

AMS sample
calculated as difference

Integrate to obtain coercivity distribution

\[ \mu_0 H_b \text{ [mT]} \]
\[ \mu_0 H_c \text{ [mT]} \]