Spin-polarized exotic nuclei:

from fundamental interactions, via nuclear structure, to biology and medicine

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UNIGE and CERN

Outline

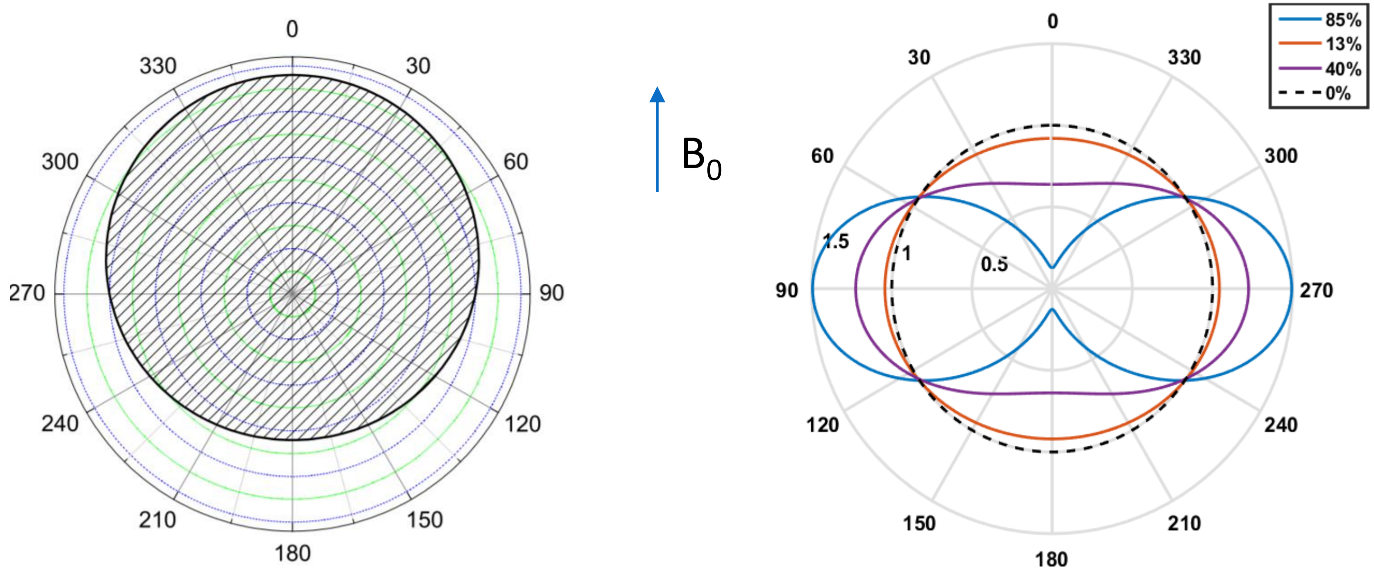
- Decay of polarized nuclei
- Spin polarization with lasers
- Spin-polarized radioactive nuclei in:
  - Fundamental physics
  - Nuclear physics
  - NMR in biology
  - MRI in medicine
- Summary and outlook

Decay of spin-polarized nuclei

- **Beta and gamma decay of spin-polarized nuclei anisotropic in space**

Beta decay, I>0

Gamma decay, I>1/2



$$W(\theta_r) = 1 + a_1 \cos(\theta_r)$$

$$W(\theta_{\mathrm{r}}) = a_0 + a_2 \cos(2\theta_{\mathrm{r}}) + a_4 \cos(4\theta_{\mathrm{r}}) + \cdots$$

$a_1 = P\, a_\beta$

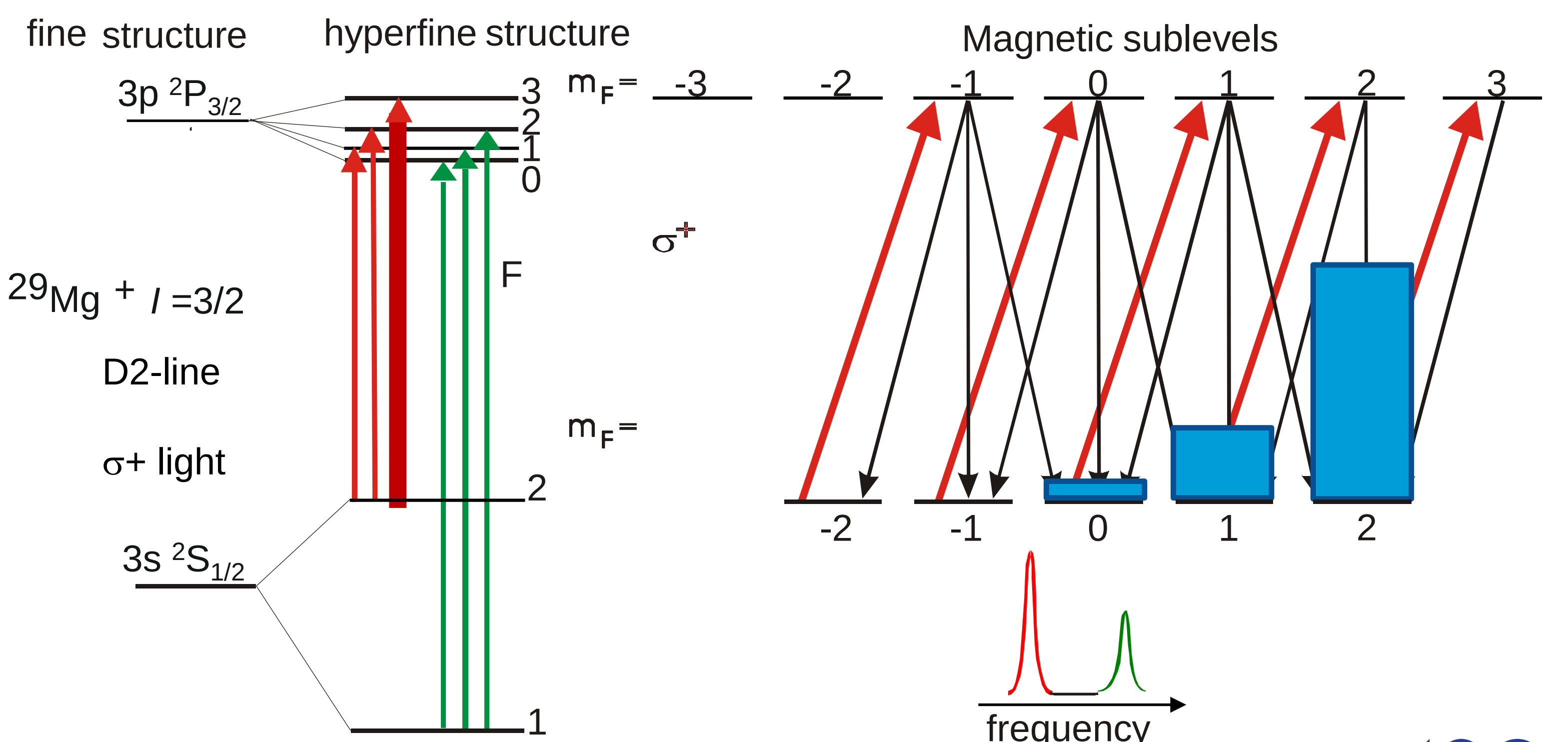
depend on degree and order of spin polarization and transition details (initial spin, change of spin)

- Observed decay asymmetry can be used to:
  - Probe underlying decay mechanism
  - Determine properties of involved nuclear states
  - Derive differences in nuclear energy levels

$$a_\beta \propto \begin{cases} 1 & \text{for } I_f - I_i = -1 \\ \frac{1}{I_i+1} \text{ (GT)} & \text{for } I_f - I_i = 0 \\ -\frac{I_i}{I_i+1} & \text{for } I_f - I_i = 1 \end{cases}$$

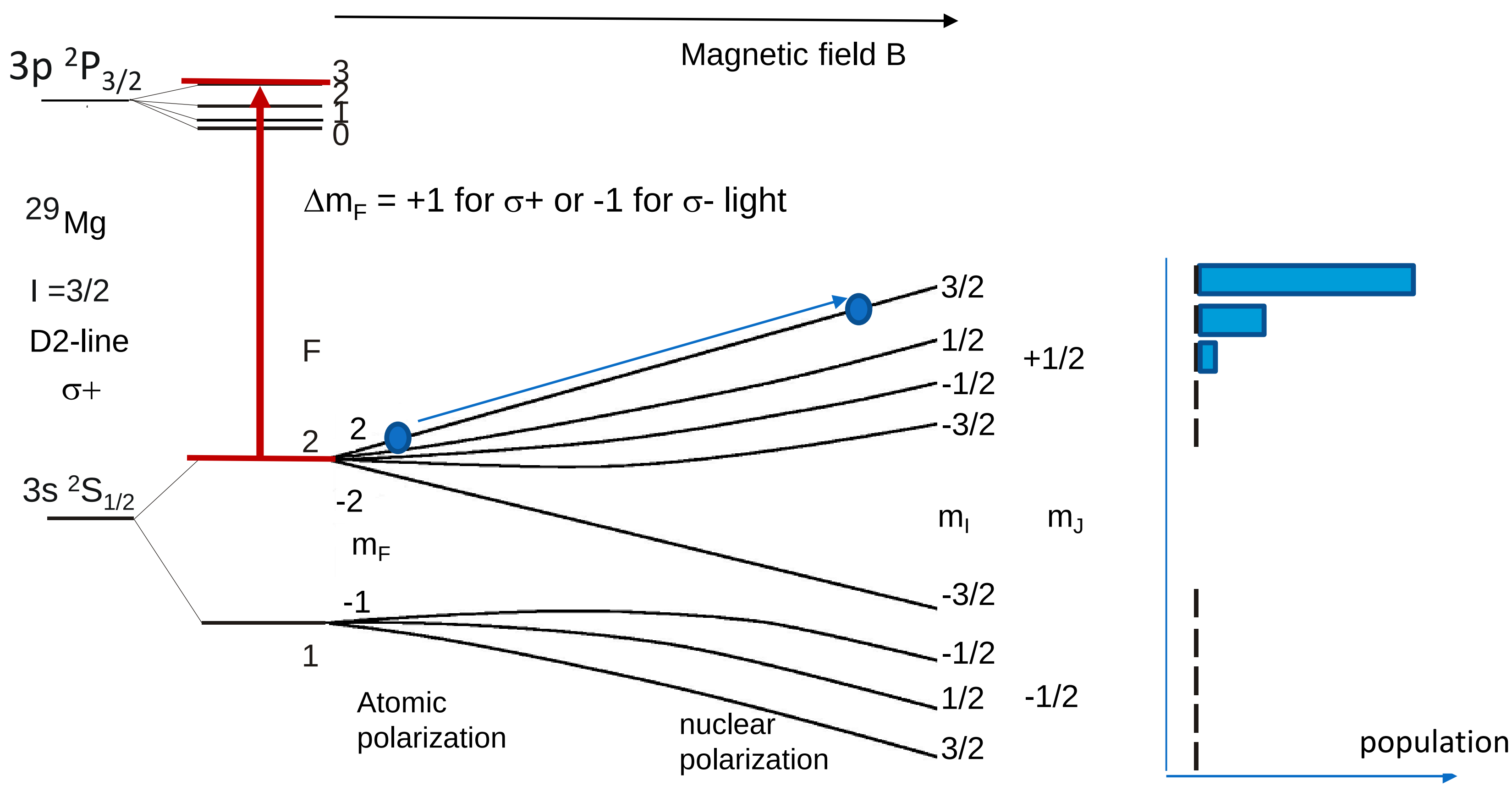
Spin polarization via optical pumping

- Multiple excitation cycles with circularly-polarized laser light
- Photon angular momentum transferred to electrons and then nuclei
  - Works best for 1 valence electron
  - nuclear spin-polarization of 10-90%
  - Polarization buildup time < us



Optical pumping and nuclear spins

- Polarization of atomic spins leads to polarization of nuclear spins via **hyperfine interaction** (P_F->P_I)
- Decoupling of electron (J) and nuclear (I) spins in strong magnetic field
- Observation of nuclear spin polarization P_I possible



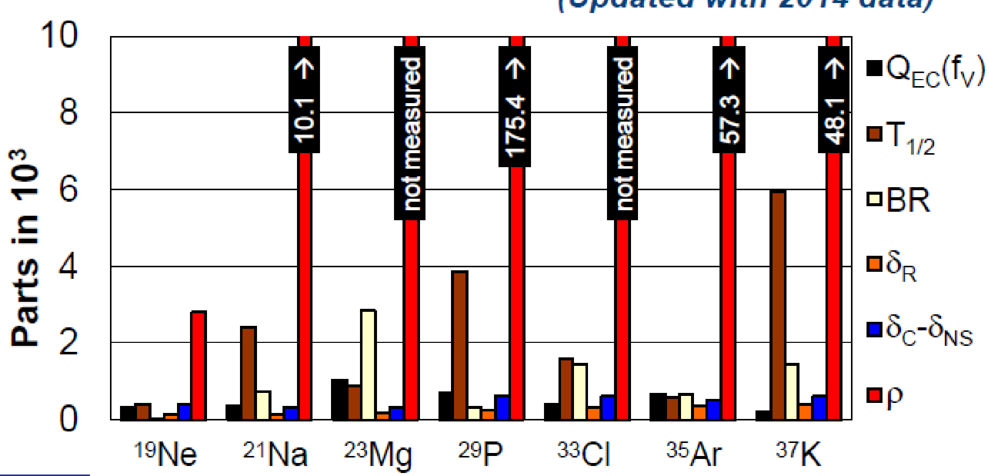
Decay asymmetry

Fundamental physics: measure V_{ud}

- V_{ud}: 1st matrix element of CKM quark mixing matrix;
- Determined from :
  - superallowed beta decays I = 0+-> 0+ and mirror isospin 1/2 decays
  - Neutron lifetime; Pion beta decay
- Latest weighted average (PDG 2018, 14 0+->0+ decays): $|V_{ud}| = 0.97420(21)$
- => CKM unitarity: $|V_{ud}|^2 + |V_{us}|^2 + |V_{ub}|^2 = 0.9994(5)$
- V_{ud} from mirror-decays (uncertainties):

O. Naviliat-Cuncic & N.S., PRL 102 (2009) 142302

(Updated with 2014 data)



$|V_{ud}| = 0.9719(17)$

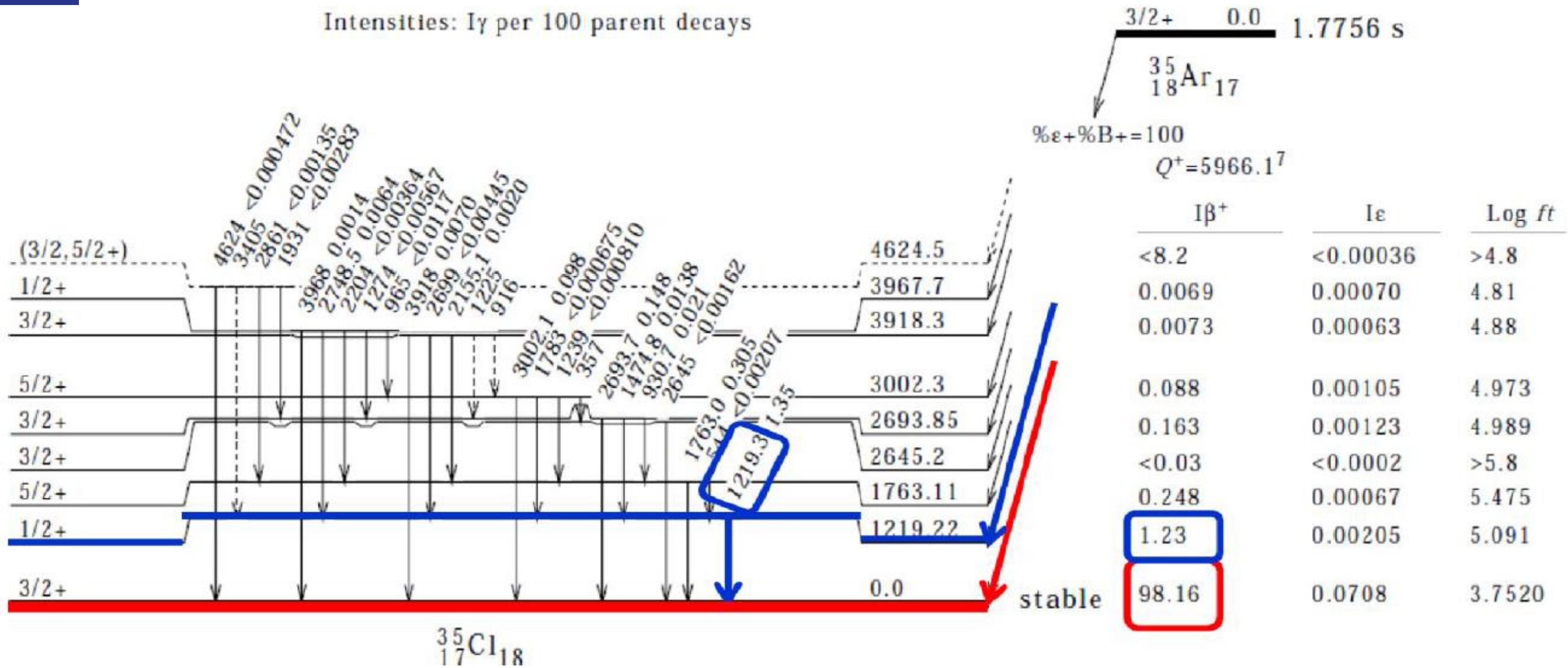
The GT/F mixing ratio: $\rho = \dfrac{C_A M_{GT}}{C_V M_F}$

- The least or **even not known quantity**!
- Precisely determined with **correlation measurements**

V_{ud} from ^{35}Ar-> ^{35}Cl mirror decay

- Measurement of 35Ar beta asymmetry factor a_β with 0.5% precision

=> Single V_{ud} value more precise than present weigthed value



a_β(gs)/ a_β(ex) = asymmetry (gs)/ asymmetry (ex) and reference a_β(ex)= 1

=> a_β(gs) = asymmetry (gs)/ asymmetry (ex) [both measured with our setup at ISOLDE]

Ph. Velten et al., *Measurement of the β-asymmetry parameter in 35Ar decay with a laser polarized beam,* Proposal to the INTC; INTC–P–426

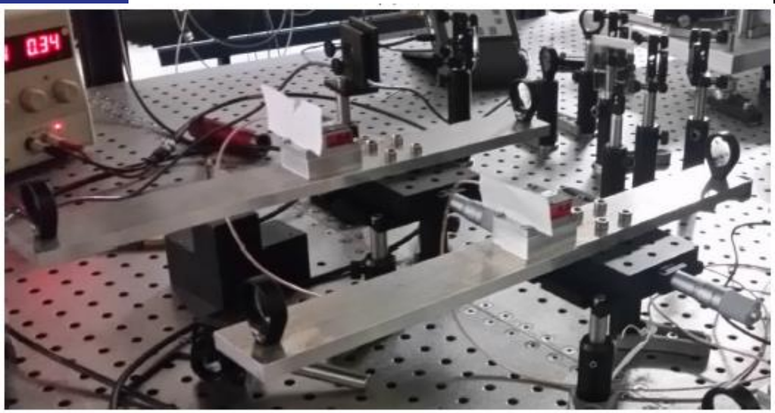
V. Gins et al, PhD thesis KU Leuven, and publication in preparation

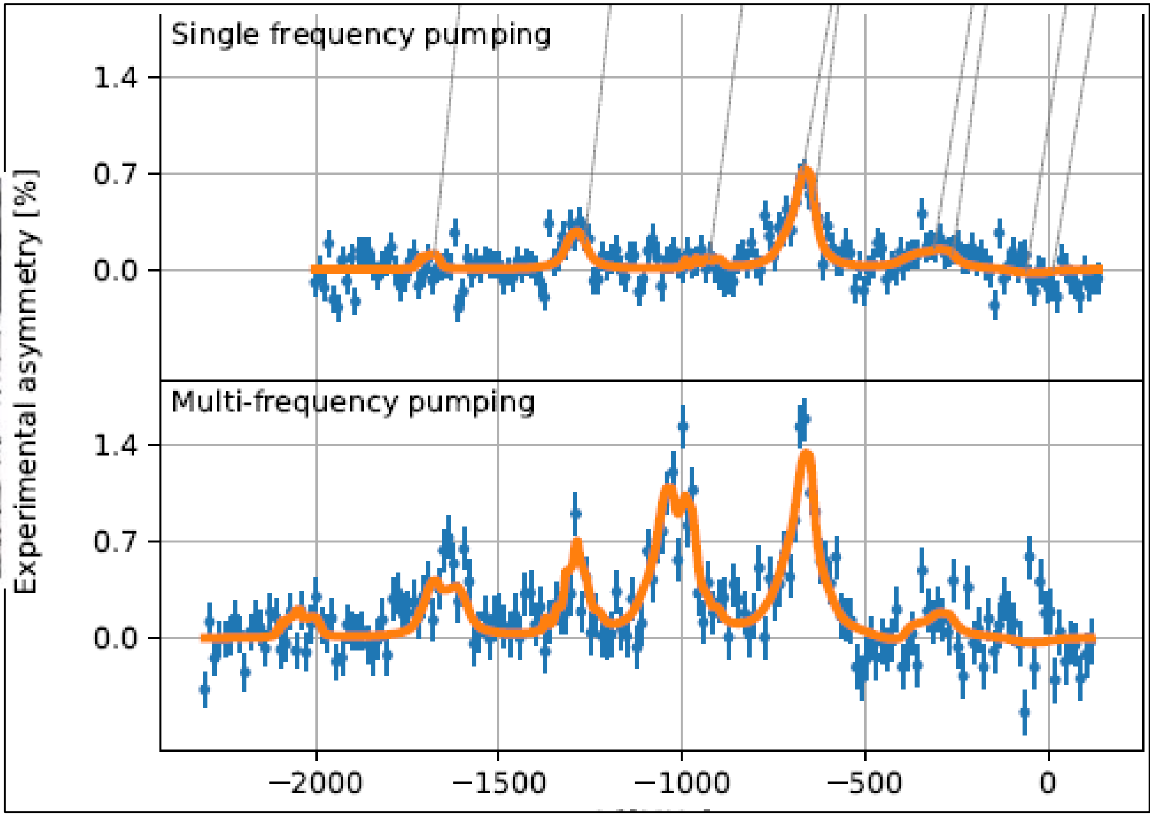
KU LEUVEN

ISOLDE

V_{ud} from ^{35}Ar decay: status

- Laser-polarization of 35Ar established and optimised
- Pumping with multiple wavelengths used for 1st time
- **max. 1.5 %** β asymmetry => too low for a_β measurement within reasonable time
- Ways of improving polarization identified (require major effort)





V. Gins et al, PhD thesis KU Leuven, and publication in preparation

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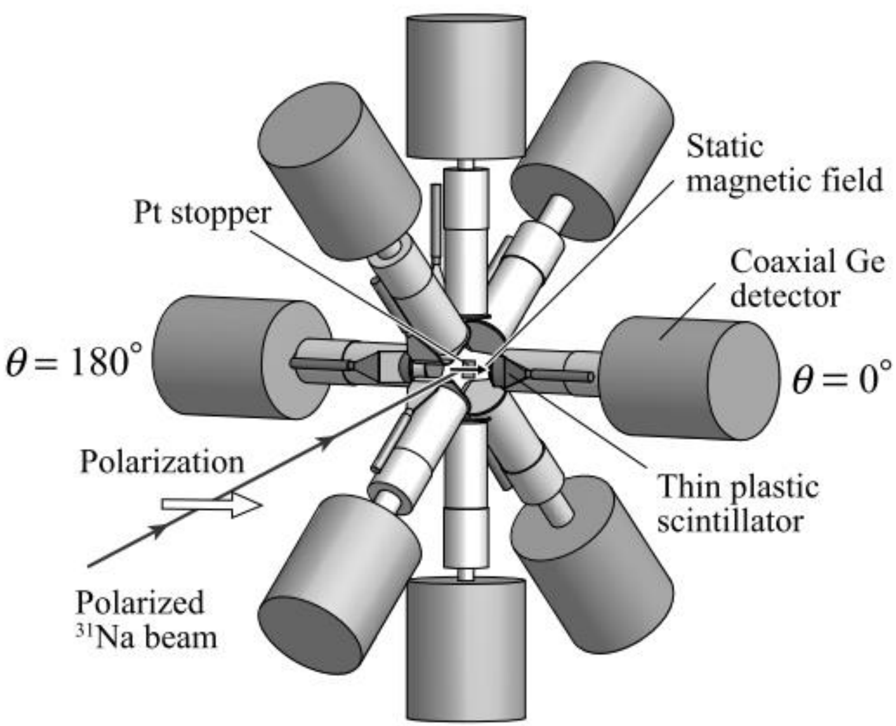
Spins/parities of states: β–γ correlations

- a_β for a given β transition depends on initial and final spins:
  - Measure angular distribution of β's in coincidence with γ's
  - Need to know degree of polarization (based e.g. on a_β = 1 transition)

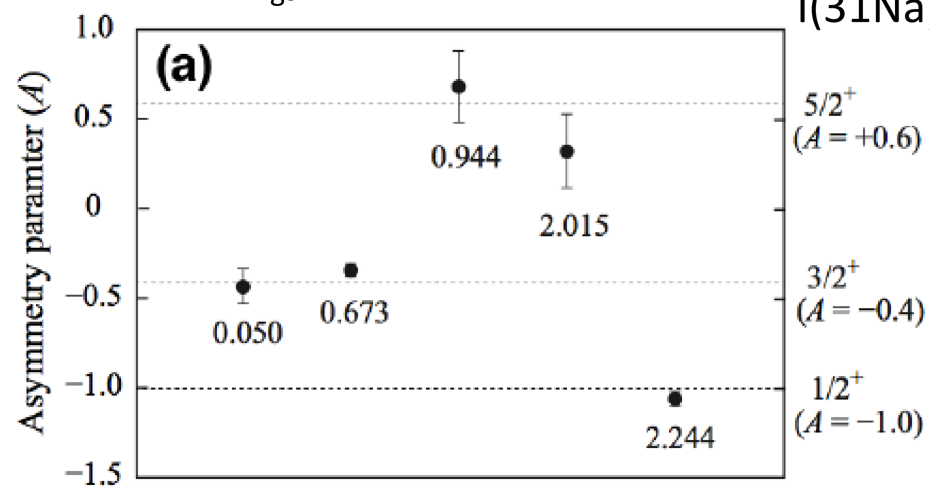
  => transition identified and a_β measured
  - One of spins known + measured a_β => other spin can be determined

- Osaka setup at TRIUMF, e.g. ^{31}Na->^{31}Mg:



I_{gs}(31Mg) = 1/2+

I(31Na)

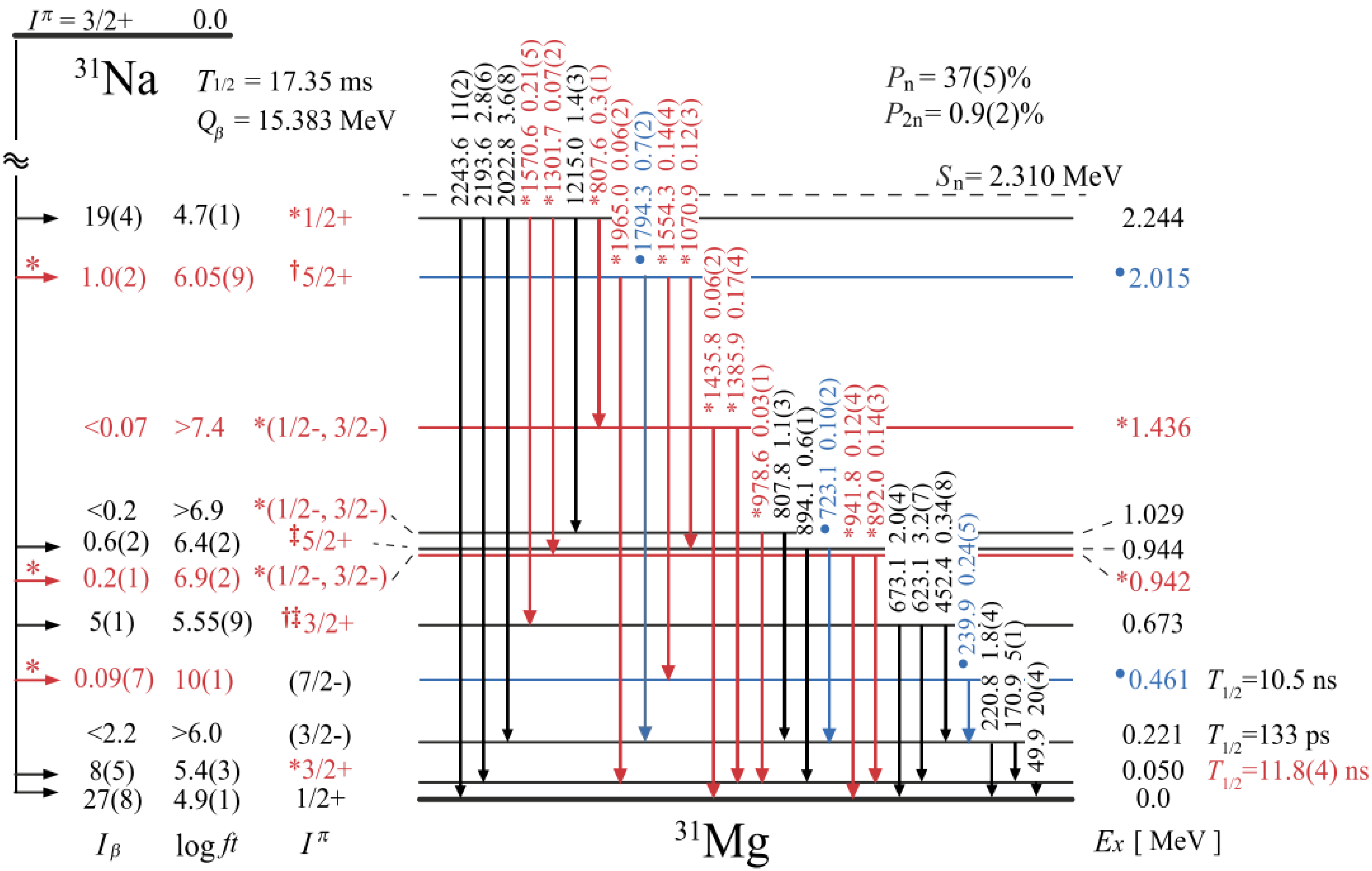


E of state emitting γ in coincidence with β's (MeV)

ɔata et al., Phys.Lett. B 767, 81 (2017).

Spins/parities of states: β–γ correlations

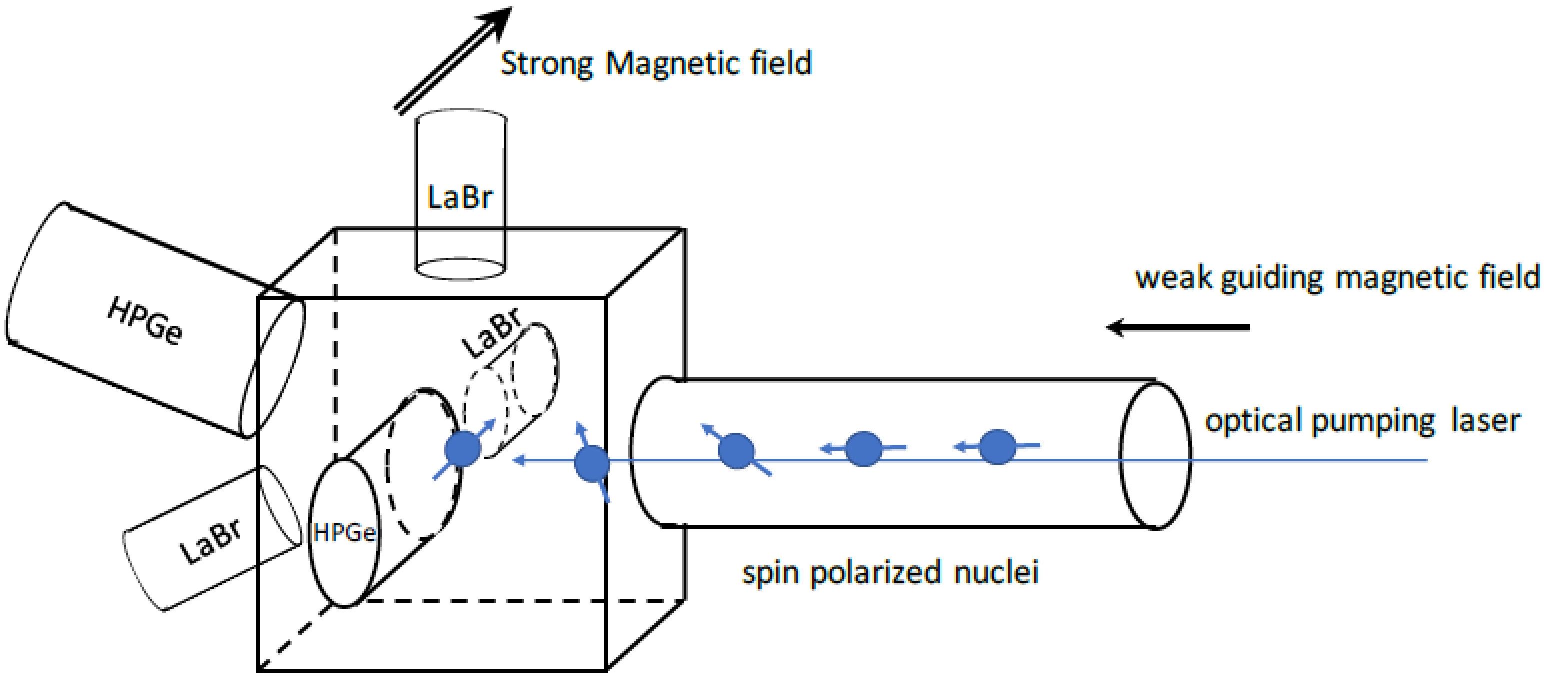
- Osaka setup at TRIUMF, e.g. ^{31}Na->^{31}Mg



H. Nishibata et al., Phys.Lett. B 767, 81 (2017).

Spins/parities with our setup

- Collaboration with Miguel Madurga, U Tennessee
- Letter of Intent submitted to ISOLDE (full proposal requested)
- To start: design of compact permanent magnets with smoothly
- Then: design of detector arrangement and their support
- Plan: setup ready when p's are back at CERN in 2021
- Later: include neutron detectors



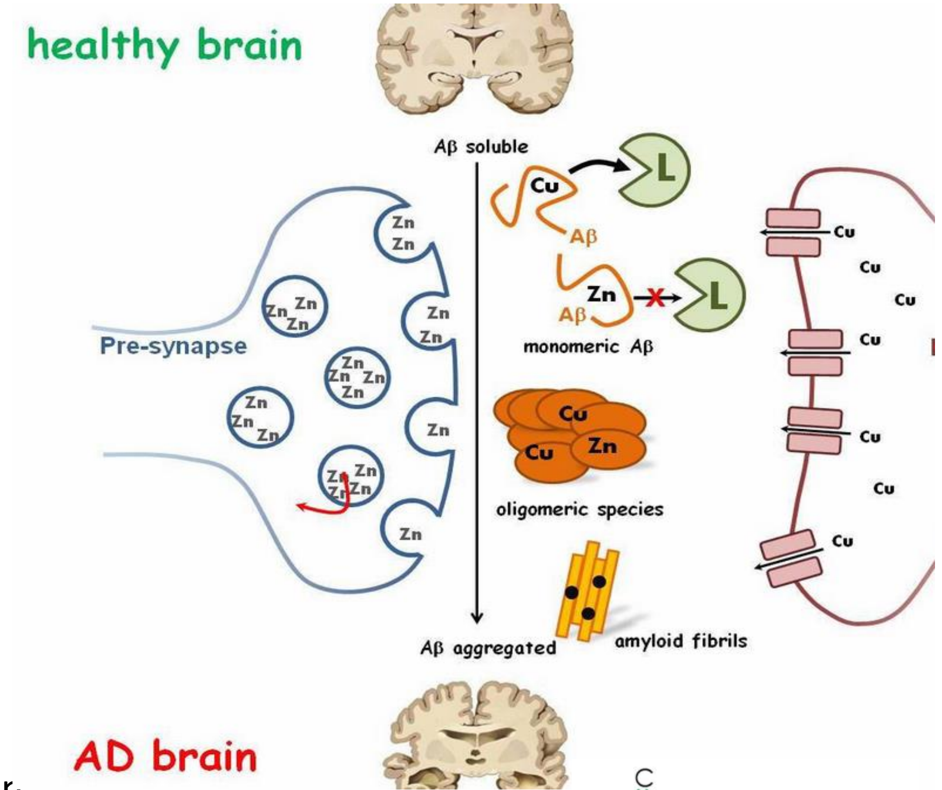
M. Madurga et al, LOI to the INTC, I-204 (2017)

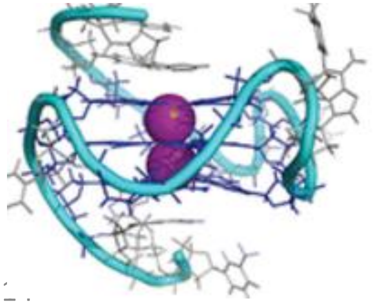
Nuclear Magnetic Resonance

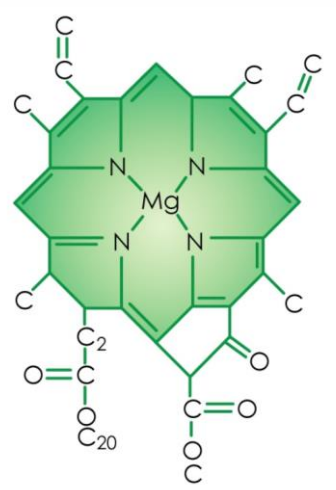
Studying metal ions in biology ?

- Role of metal ions in human body depends on adopted coordination environment

- Right concentration crucial for correct functioning of cellular processes
  - Na, K: transport of sugars and amino acids into cells; regulate flow of water across membranes
  - Mg: RNA- and DNA-processing enzymes and ribozymes
  - Cu: present in many enzymes involved in electron transfer and activation of oxygen
  - Zn: 2nd most abundant trace element in human body; catalytic and structural role, regulation of genetic message transcription and translation

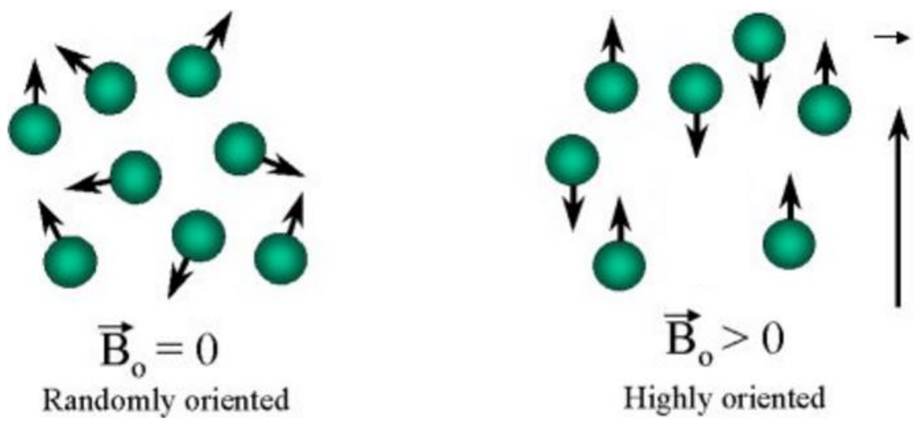


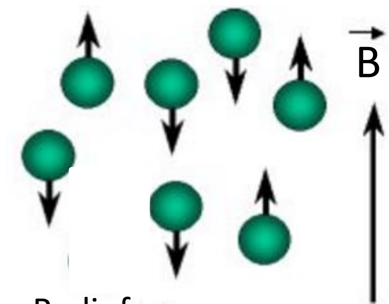




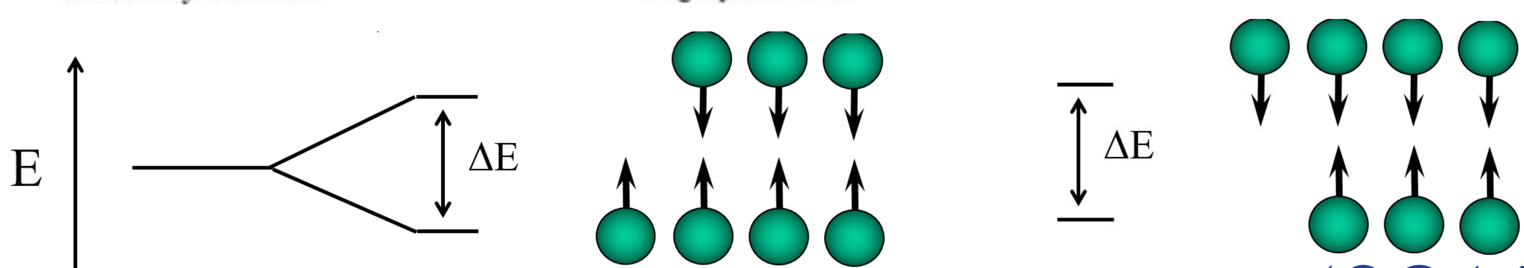
NMR principles

- Participants:
  - Probe nuclei with spin different from 0
  - Sample/ environment
- Magnetic field
  - Strong static field (B0) -> when different at different positions –> **MRI (Magnetic Resonance Imaging)**
  - Weaker field (B1) oscillating at radio-frequency (MHz)





Radiofrequency:
Change in orientation



NMR in nuclear physics

Method to determine precisely magnetic & quadrupole moments of short-lived nuclei

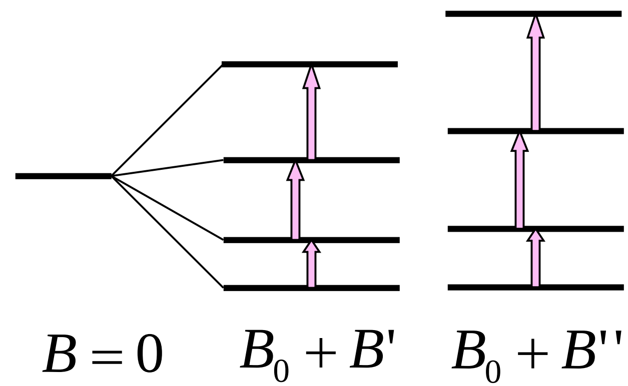
- **Observables**: Larmor frequency
- **Determined properties**
  - Magnetic dipole and electric quadrupole moment of the studied nucleus

Depends on nucleus

$$\Delta E_{mag} = |g_I| \cdot \mu_N \cdot B + \frac{1}{2} Q \cdot V_{zz}$$

known

Same nucleus, different host



- **Derived information**:
  - Magnetic moment – orbitals occupied by valence nucleons
  - Quadrupole moment – collective properties

- Example: spin and magnetic moment of 31Mg
  - M. Kowalska, PhD Thesis, U Mainz 2006

NMR in (chemistry and) biology

Most versatile method to study structure and dynamics of molecules in solution

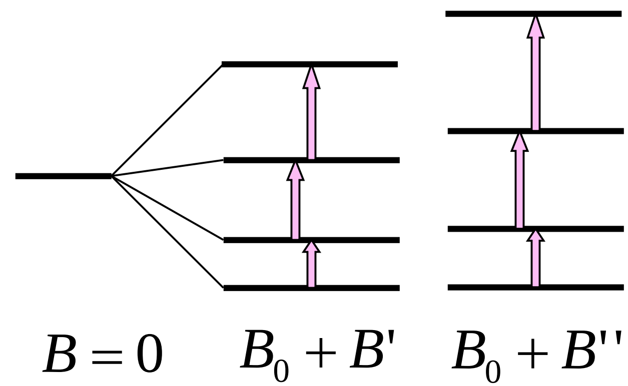
- **Observables**: chemical shift (Larmor frequency) and relaxation times in different hosts
- **Determined properties**
  - ➢ local electronic environment (i.e. **number and type of coordinating groups**)

Depends on environment

$$\Delta E_{mag} = |g_I| \cdot \mu_N \cdot B + \frac{1}{2} Q \cdot V_{zz}$$

known

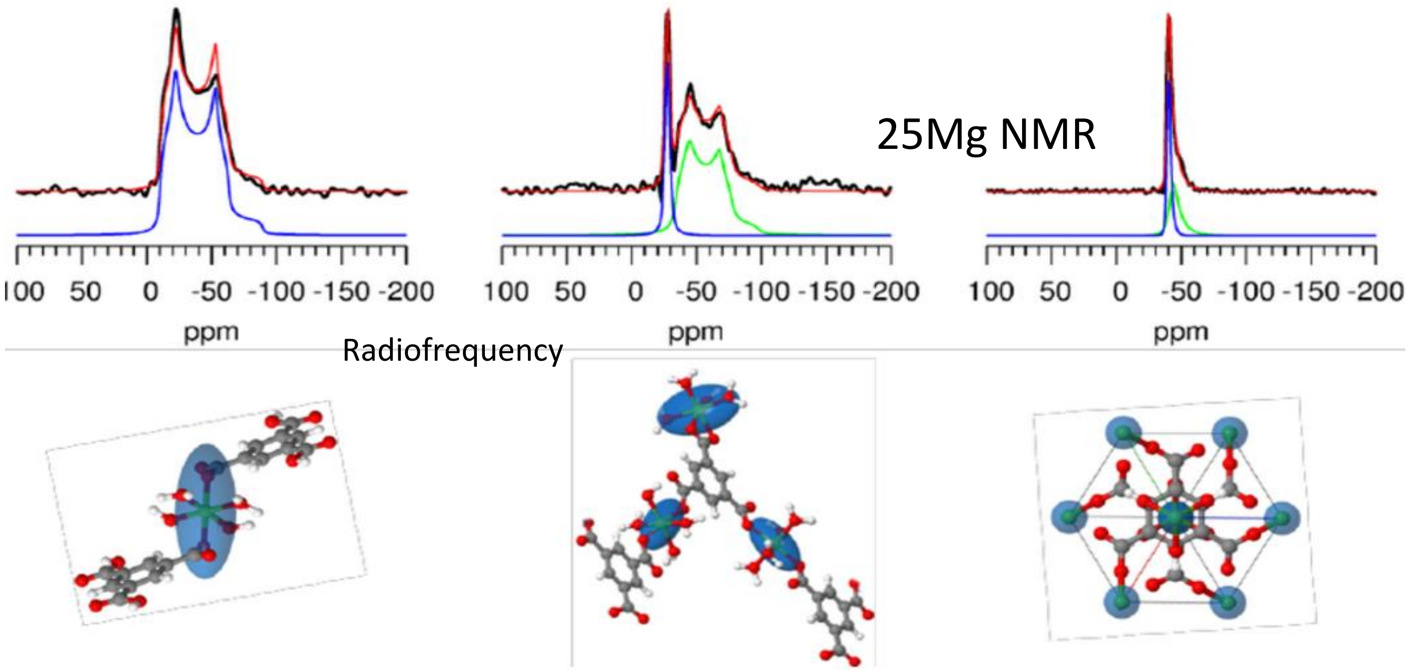
Same B, different shielding by host



- **Derived information**: comparison to quantum-chemical models (e.g DFT)
  - ➢ kinetics and dynamics and ligand binding of the **metal ions and biomolecules**
  - ➢ 3D structure of proteins and **protein-metal complexes**

Metal ions & NMR

- NMR can provide info on location & evolution of metal-ion binding to biomolecules
  - NMR on 23Na, 25Mg, 63Cu, 67Zn bound to the biomolecule



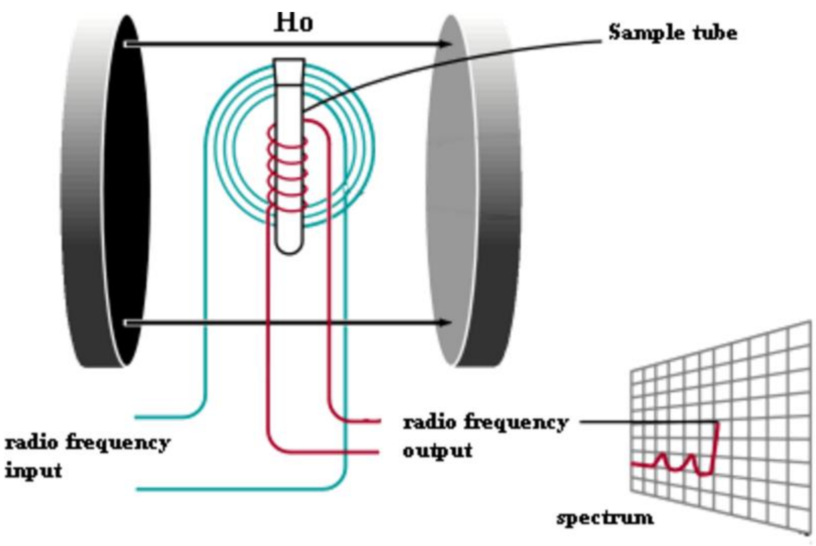
- Challenges:
  - almost invisible signals due to small abundance, I>1/2, and small sensitivity (due to small magnetic moment)

- In common with radioactive nuclei:
  - Small amount of nuclei so a sensitive NMR approach is needed

NMR limitation: sensitivity

- NMR is powerful but not sensitive
  - Small degree of polarization
  - Inefficient detection

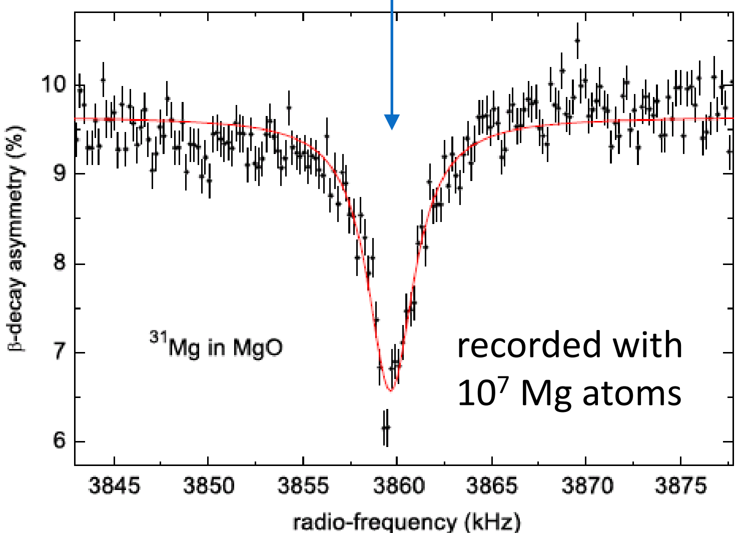
- Our combined paths to increase sensitivity: **beta-NMR and gamma-NMR/MRI**
  - Hyperpolarization -> optical pumping with lasers
  - Detection of particles -> asymmetry in beta or gamma decay

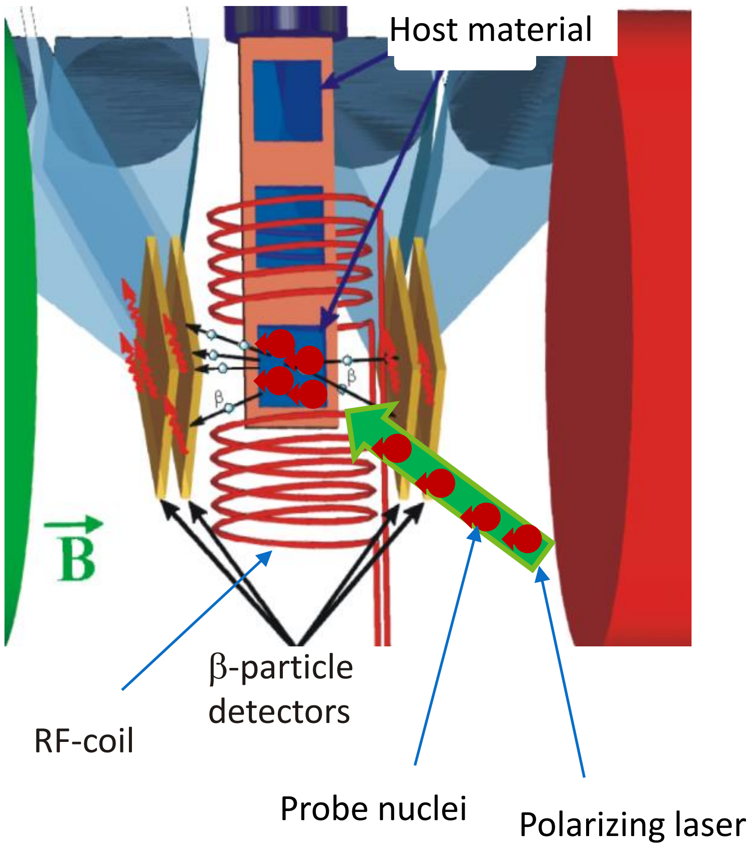


Beta(-detected) NMR

- Same principles as conventional NMR
- Ingredients:
  - Radioactive (short-lived) NMR-active beta-decaying nuclei **brought from outside**
  - Beta particles emitted in spin direction
- Detection of resonance:
  - **Asymmetry in beta decay in space**
  - **At resonance: decrease in asymmetry**
- When combined with hyperpolarization

=> Beta-NMR can be up to 10^{10} more sensitive than conventional NMR

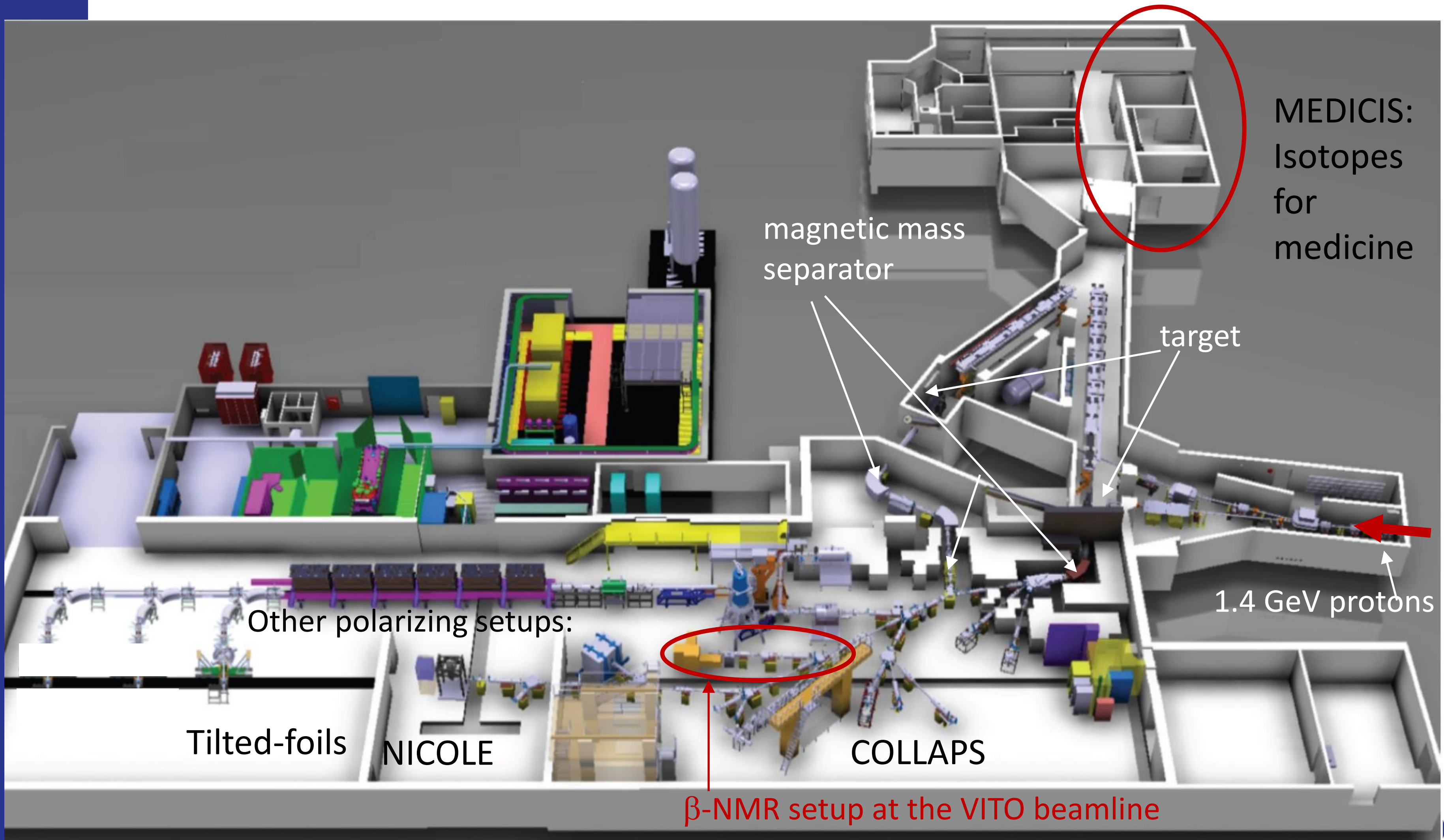




M. Kowalska, PhD thesis

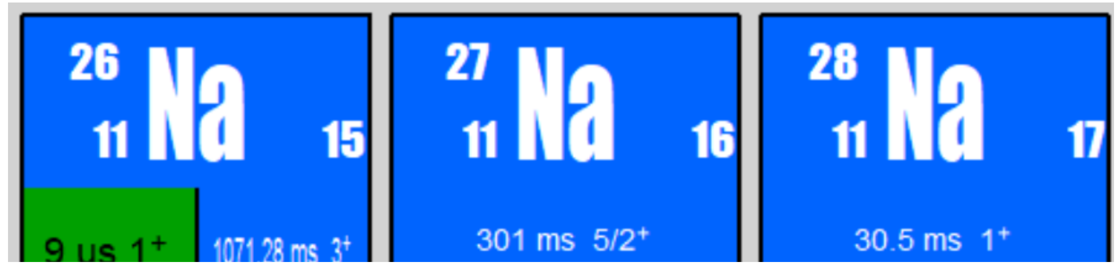
Spin-polarization & β-NMR at ISOLDE-CERN

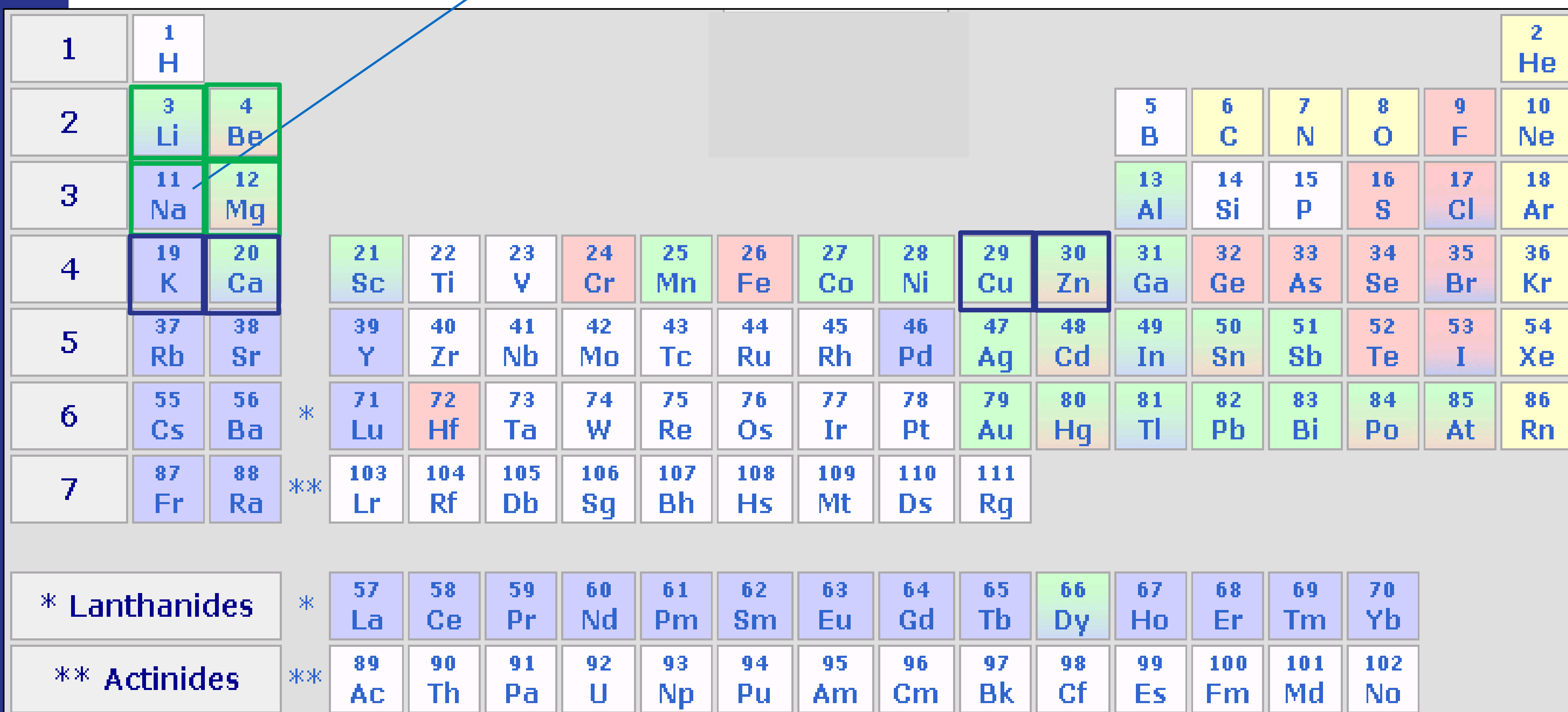
Laser-polarization and β-NMR at VITO beamline



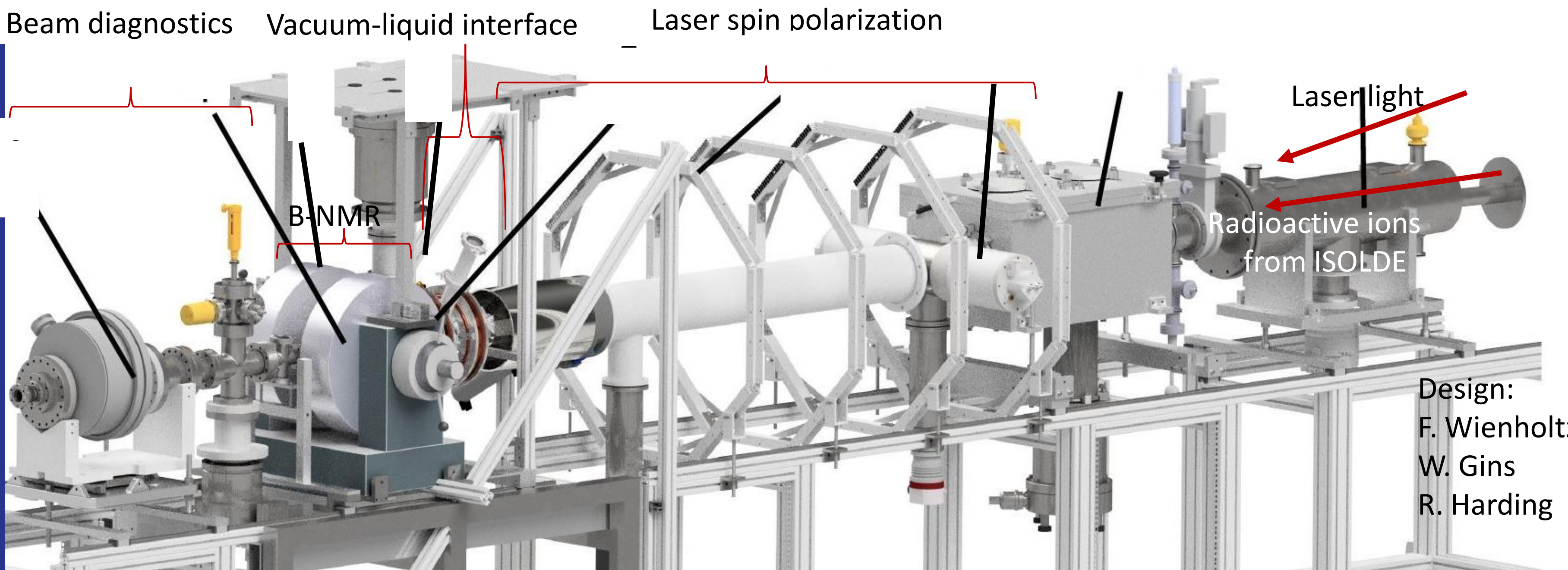
ISOLDE selection of radio-nuclei

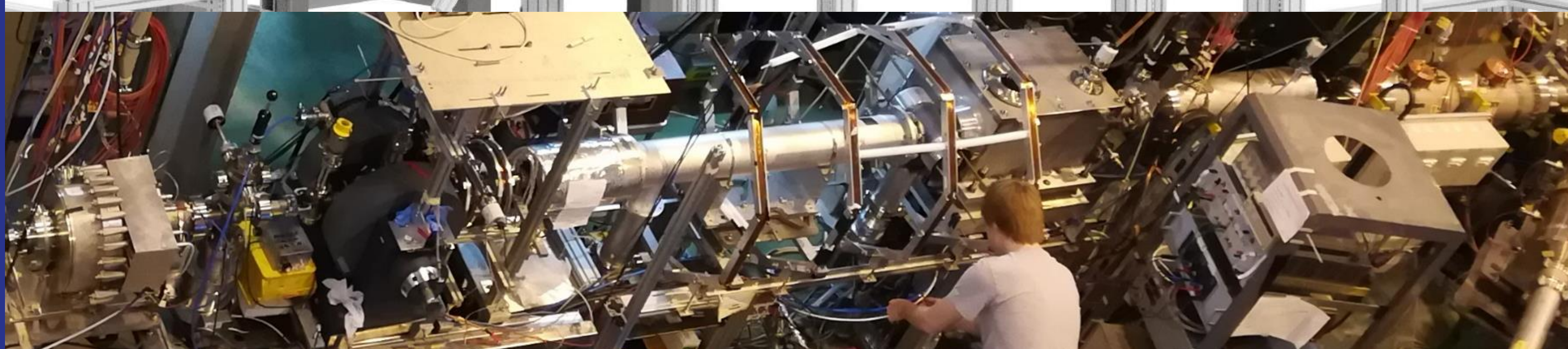
- Over 1200 isotopes from 80 chemical elements used for experiments
  - Radioactive half-lives: >10 ms
- Laser-polarized metals:
  - Already polarized at ISOLDE
  - Planned within this project





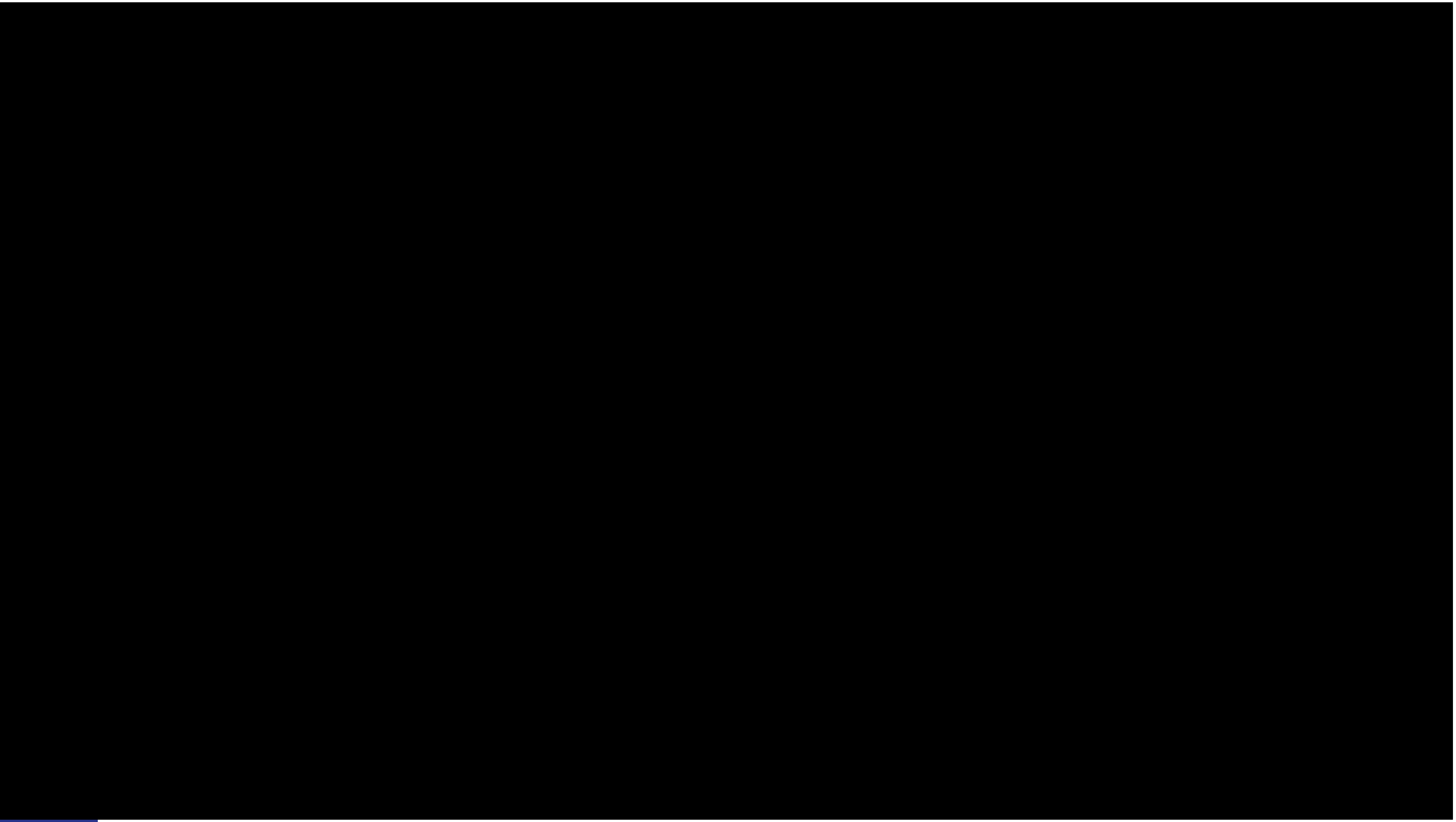
Experimental setup



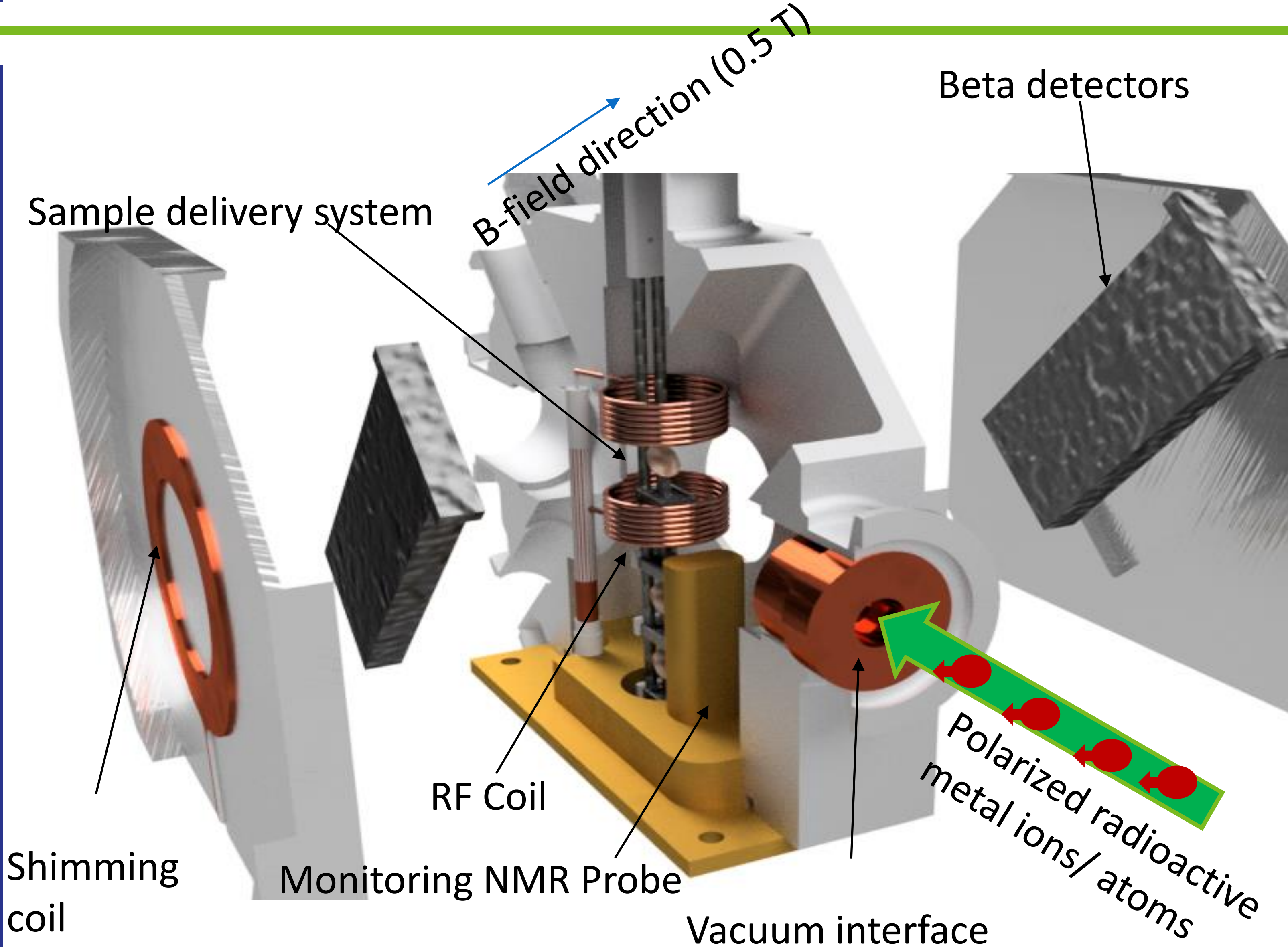


M. Kowalska et al., J. Phys. G: Nucl. Part. Phys. 44 (2017) 084005
W. Gins et al., submitted to Nucl. Instr. and Meth. A (2018)
W. Gins, PhD Thesis, KU Leuven (2019)

Designed and commissioned in 2016
First physics experiments in 2017
First biology-related experiments in 2018

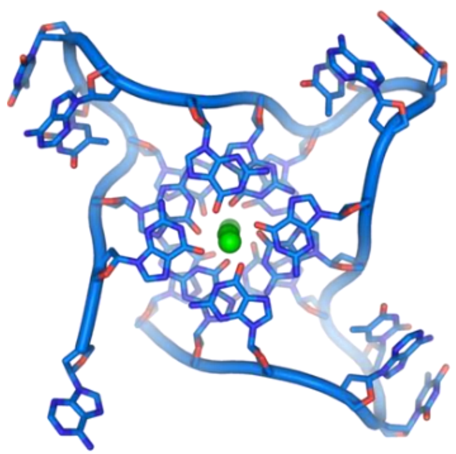


Bio-Beta-NMR chamber

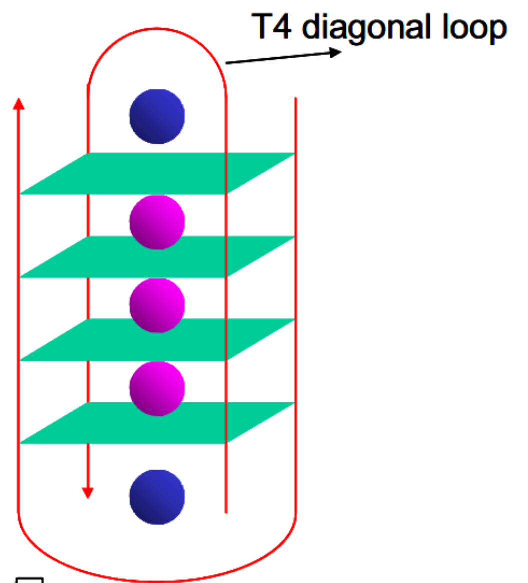


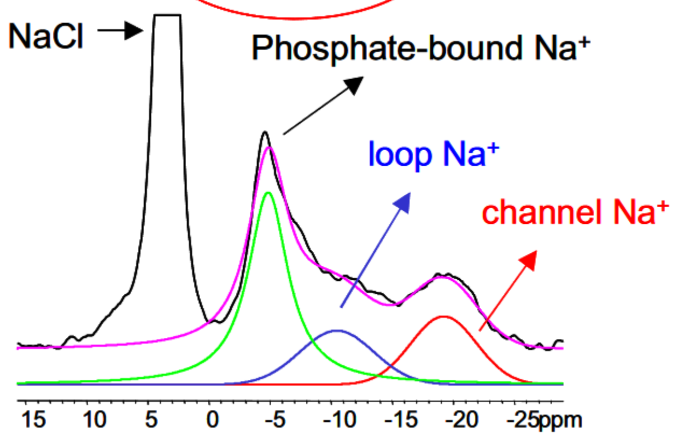
First bio-study: Na+ & G-quadruplexes

- DNA G-quadruplexes:
  - Guanine-rich DNA fragments
  - Found in nature, e.g. in telomeres or oncogenes
  - Synthesised for novel applications
  - Important in different diseases



One of few 23Na NMR GQ studies:
R. Ida, G. Wu, JACS, 2008



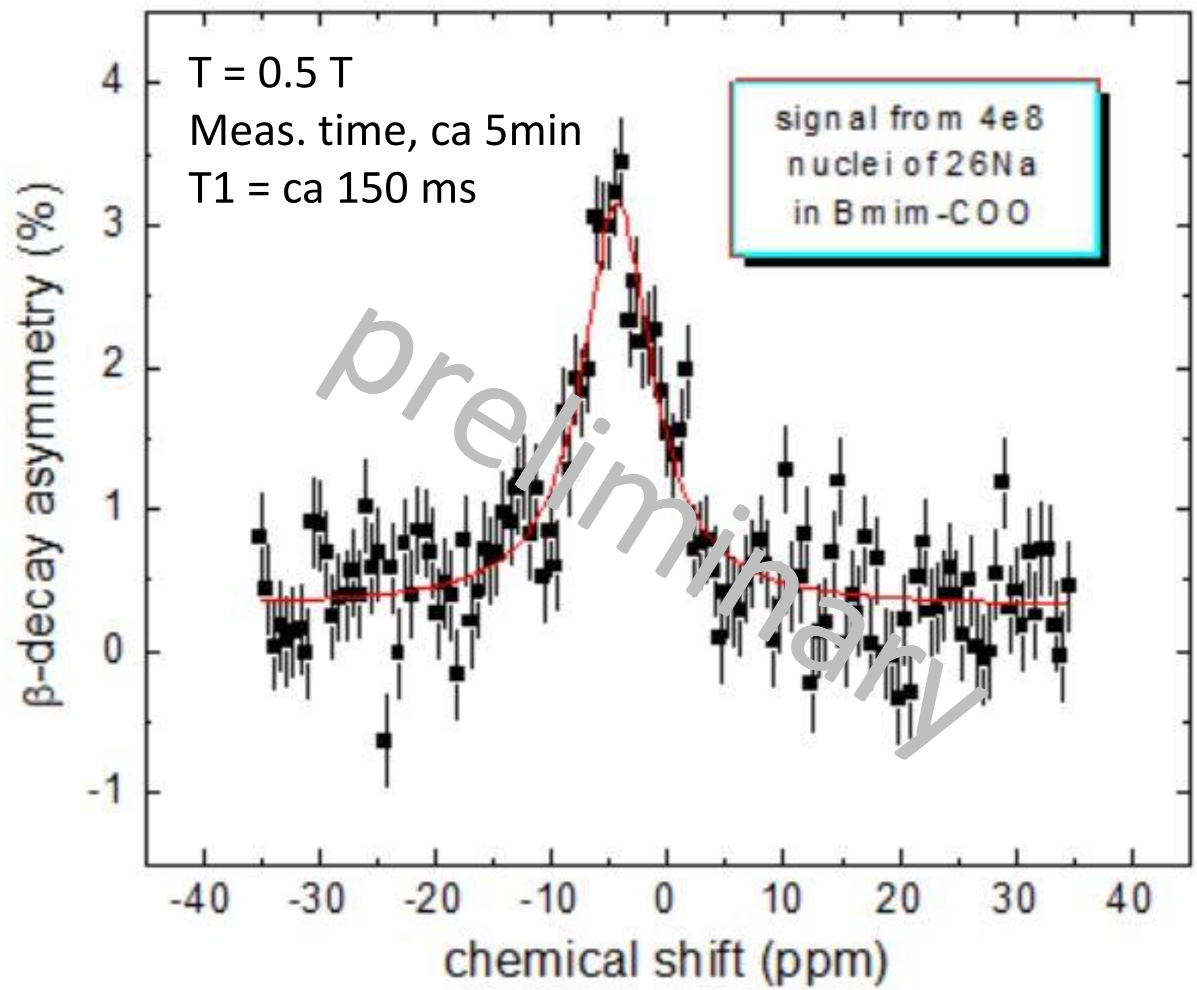


- Alkali metals in DNA G-quadruplexes
  - Important for their formation, stability and structural polymorphism
  - Until recently considered invisible in conventional Na+/K+ NMR

M. Kowalska et al, Proposal to the ISOLDE Scientific Committee, June 2017
M. Trajkovski et al, J. Am. Chem. Soc. 134, 4132 (2012)

1st liquid beta-NMR results

- Dec 2017: First Na beta-NMR signals in liquid hosts

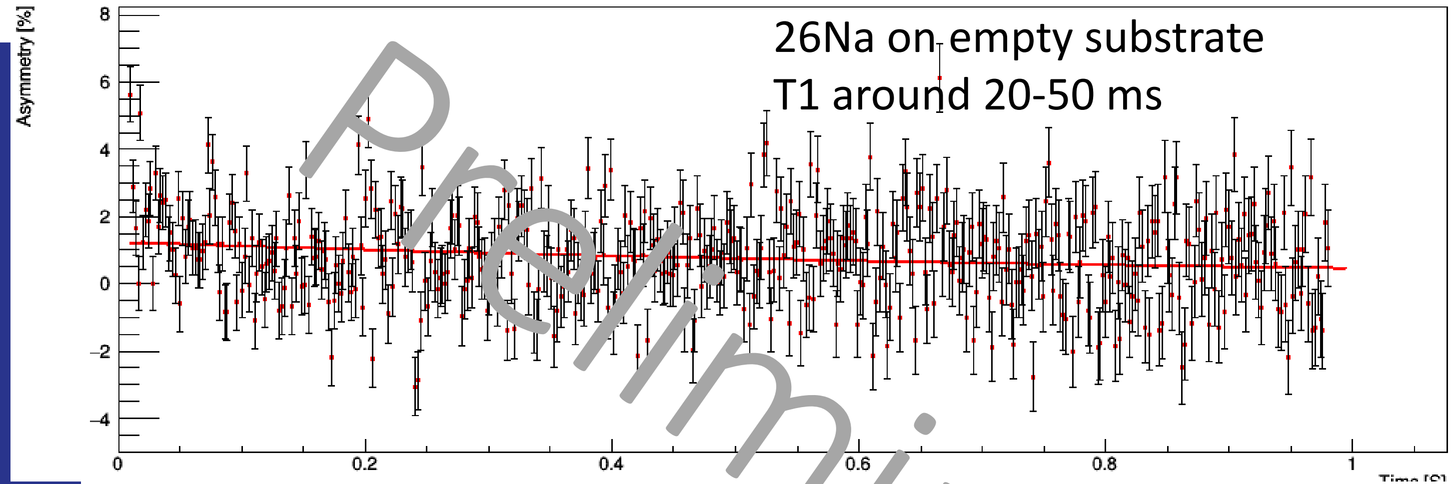


26Na
T1/2 = 1.1 s
I = 3
μ = 2.86 μ_N
Q = - 5 mb

In comparison - 23Na:
I = 3/2
μ = +2.21 μ_N
Q = 100 mb
(20x larger!)

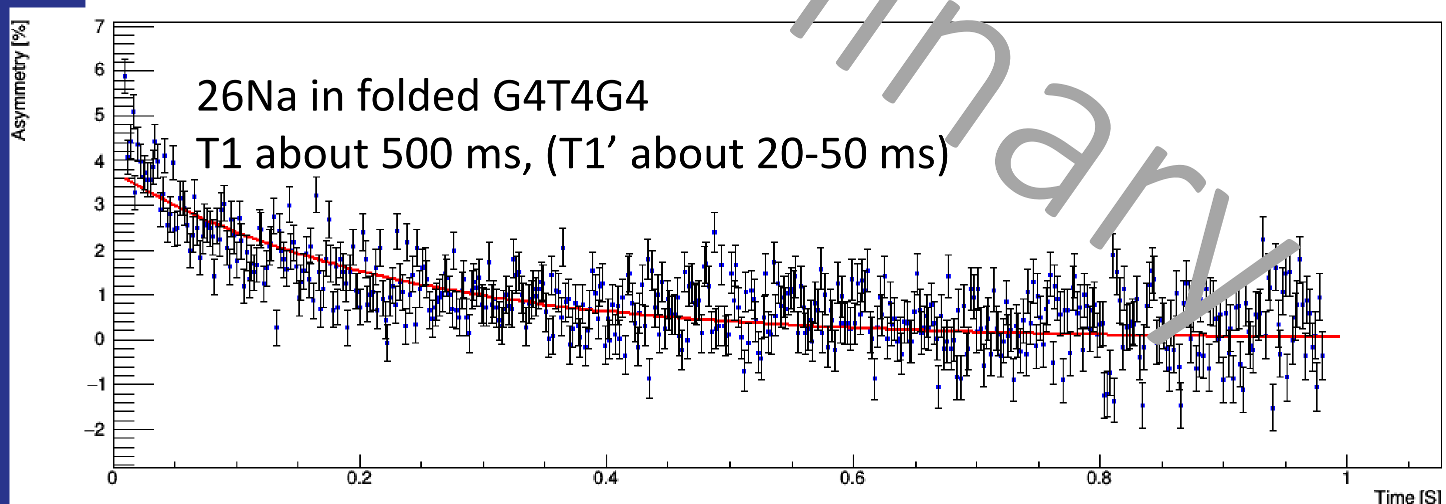
Resonance width will allow determining chemical shifts expected for Na

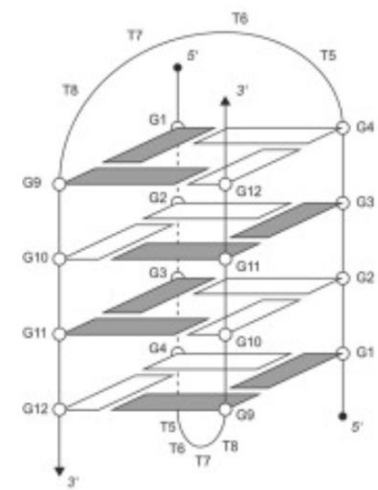
β-NMR: 26Na T1 in G4T4G4 G-quadruplex



May 2018

Folded G4T4G4 DNA sequence on substrate



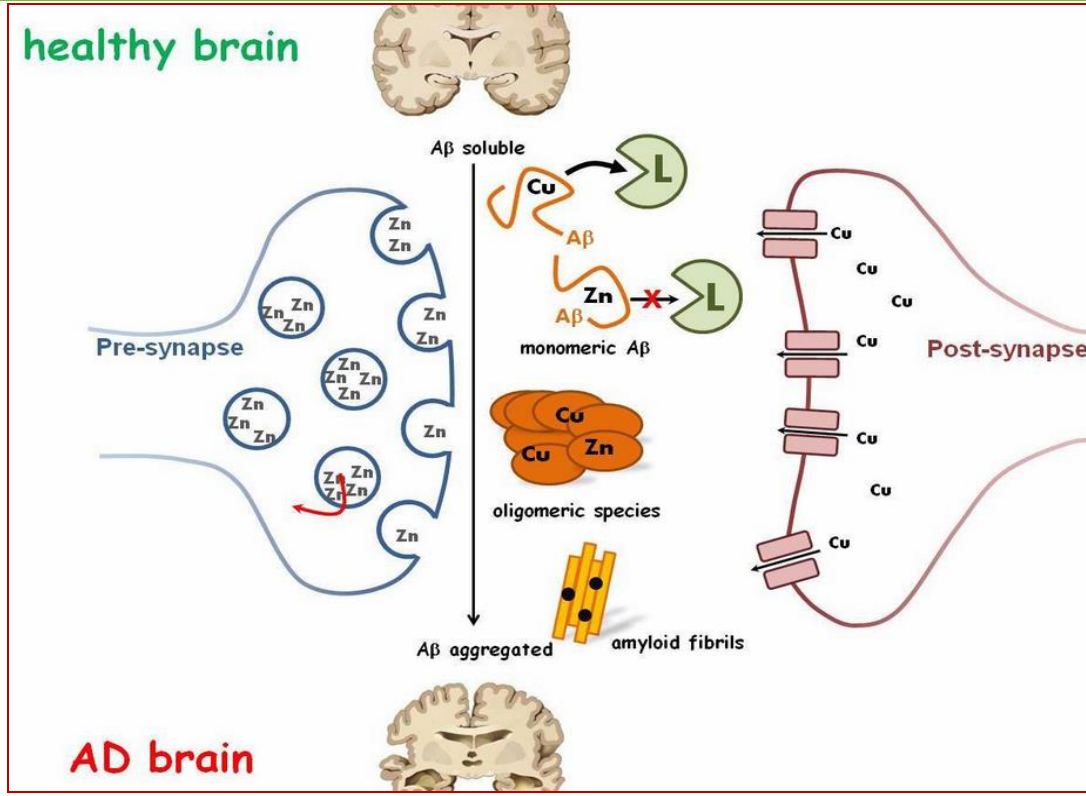


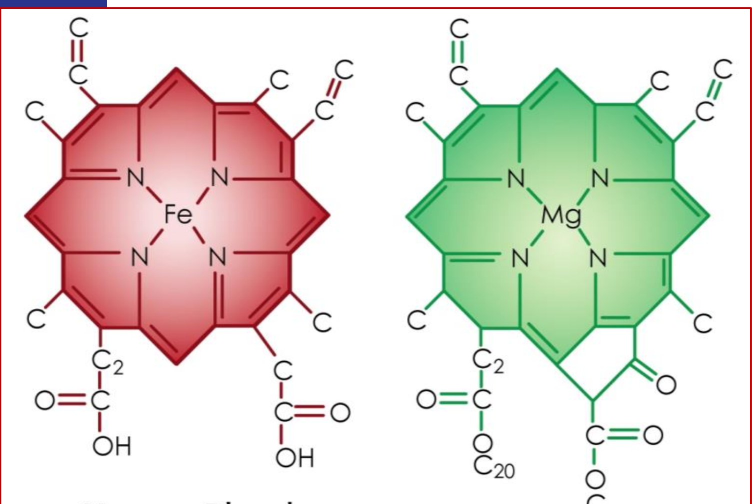
- T1 in presence of GQ quite long (due to relatively symmetric environment of GQ?)
- Calculations should help in more detailed interpretation
- Oct18 NMR spectra under analysis – probably too broad to see -17 ppm shift

Beyond 2018

Already polarized at ISOLDE:
8,9Li
11Be
26-28Na
29,31Mg

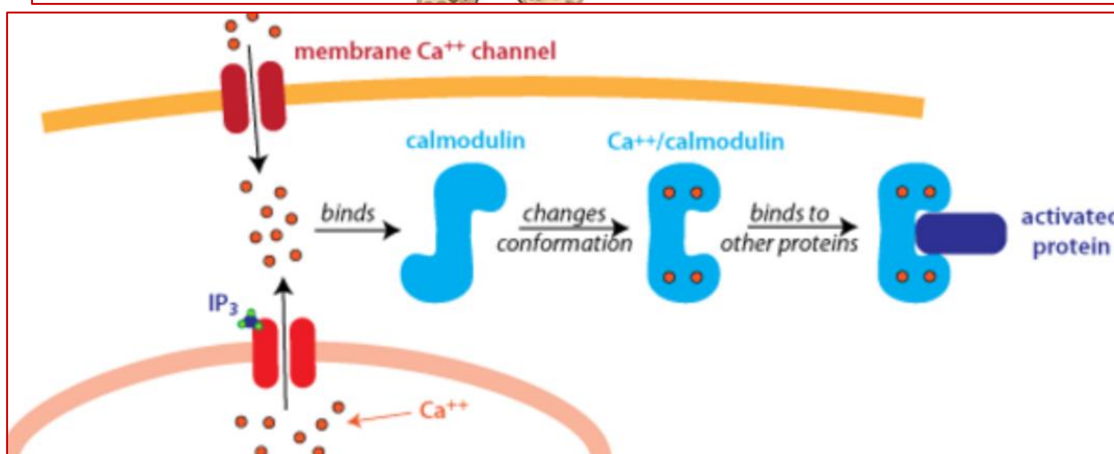
Feasible and planned soon:
37,49K
39,51Ca
58,74,75Cu
75,77Zn





Human Blood Hemoglobin

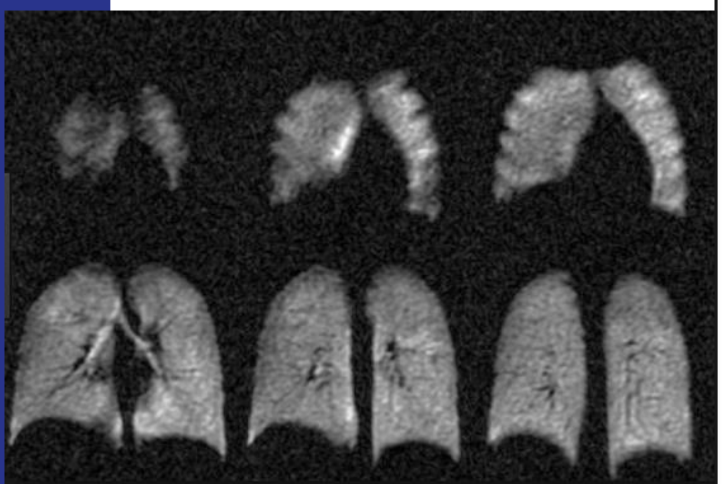
Plant Chlorophyll

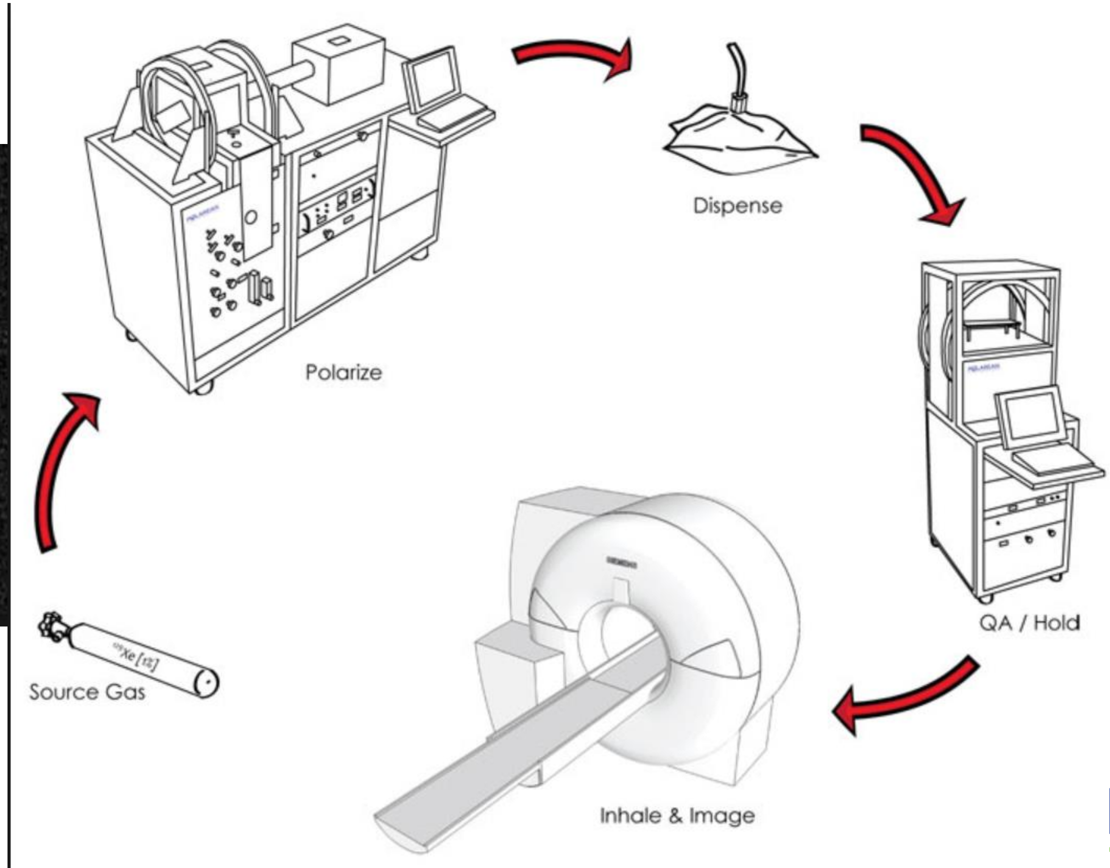


Magnetic resonance imaging

MRI with hyperpolarized nuclei

- MRI: high resolution, but small sensitivity
- Hyperpolarization: increase in sensitivity by up to 1e5
- Best example: 129Xe:
  - Polarized via spin-exchange with laser-polarized Rb
  - Applications: lung and brain MRI, encapsulation and use in body liquids





1st medical applications of 3He:
W. Heil et al, Mainz, Nature 1996

Proposed new modality: gamma-MRI

PET/SPECT and MRI have complementary features:

	Detection efficiency	Spatial resolution
PET and SPECT	High (PET>SPECT)	Low (e.g. >5mm for 82Rb)
MRI	low	High

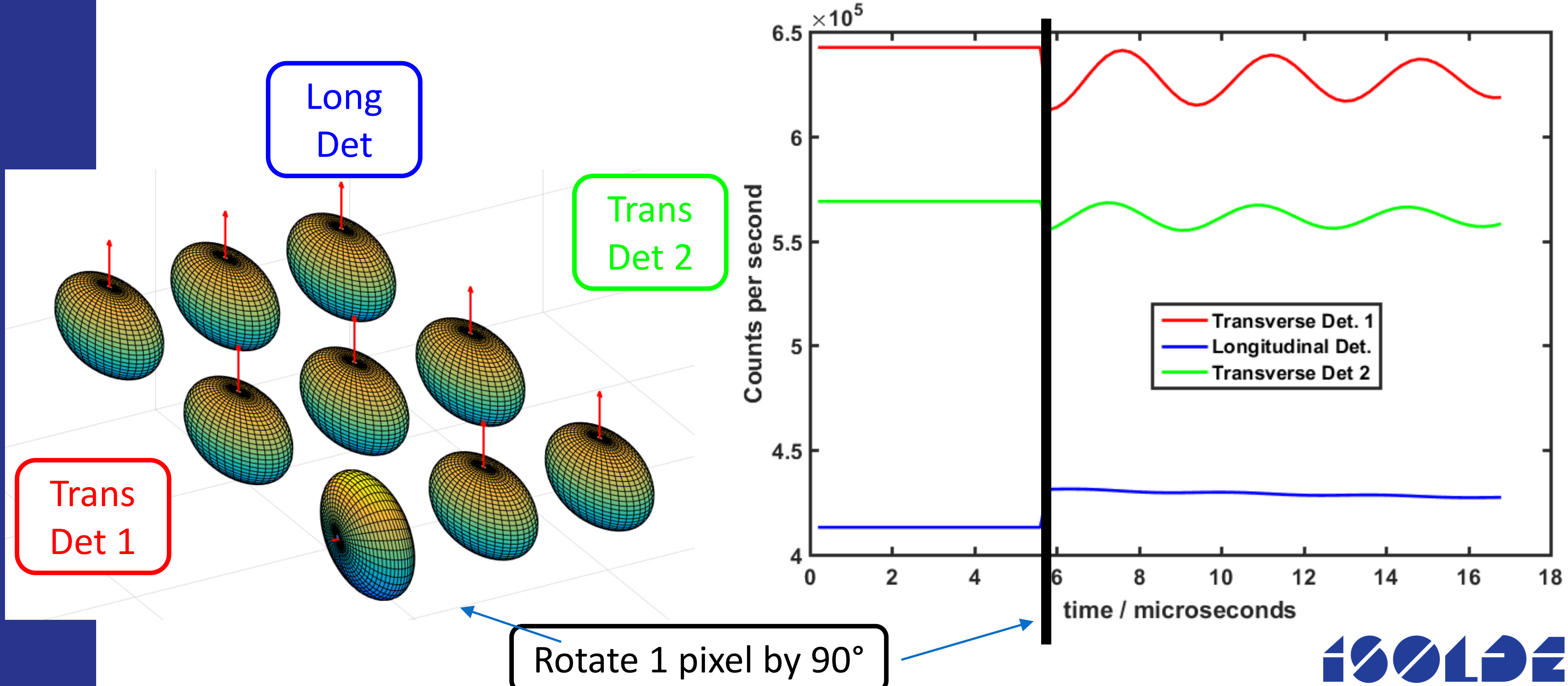
Solution: **gamma-MRI (or simultaneous SPECT-MRI):**

- What - Record MRI signals from PET/SPECT-type nuclei
- How - Hyperpolarize spins and observe asymmetry of gamma decay
- Result - high efficiency (gamma detection) and high resolution (MRI)
- Status: method shown to work by US team: Y. Zheng, et al., Nature 537, 652 (2016)

- Gamma-MRI Equipment:
  - I>1/2 gamma-emitting nuclei
  - Spin-polarizer
  - MRI magnet
  - **Gamma detectors inside B field**

Gamma MRI – spatial resolution

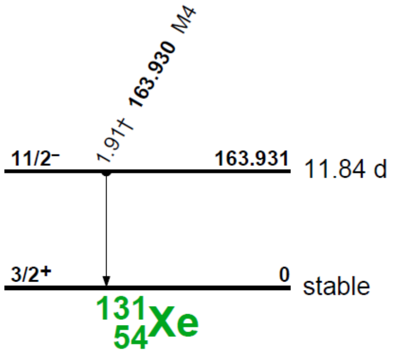
- Voxel size
  - defined by slope of B-field gradients and spectral width of rf pulse
  - more nuclei -> smaller pixels possible up to B gradient and rf limit
- 1 pixel in resonance:
  - change in total gamma counts visible in each detector
  - Degree of change proportional to number of nuclei in addressed pixel

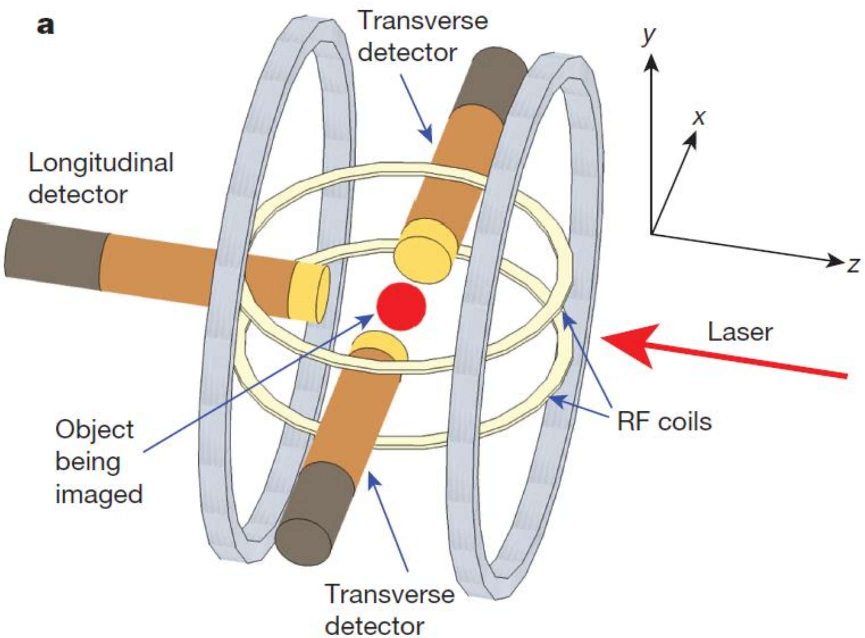


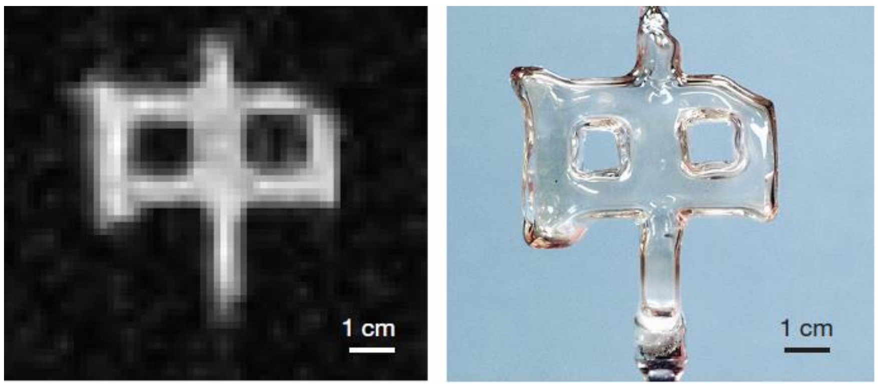
First gamma-MRI

Y. Zheng, G.W. Miller, W.A. Tobias, G.D. Cates, Nature 537, 652 (2016)

- **131mXe**: t1/2 = 12 days
- **Setup:** low B-field
- **Results:** space-resolved signal (recorded pixel after pixel) with 1e13 nuclei vs 1e24 normally

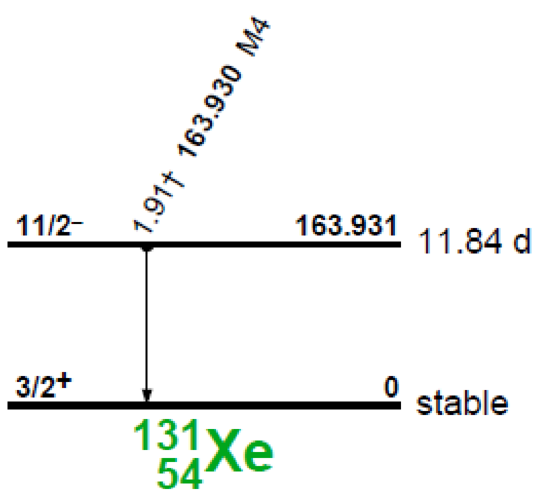


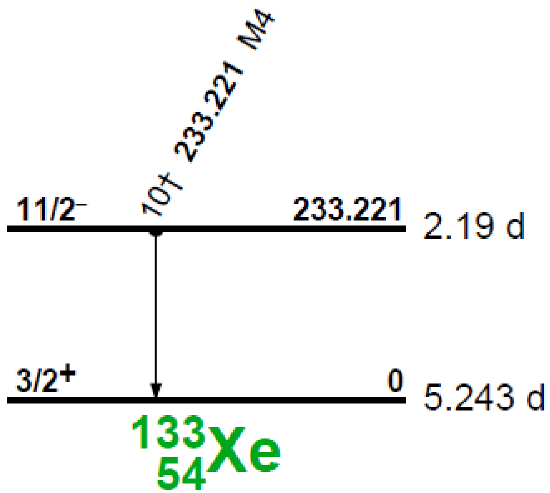




Our gamma-MRI project

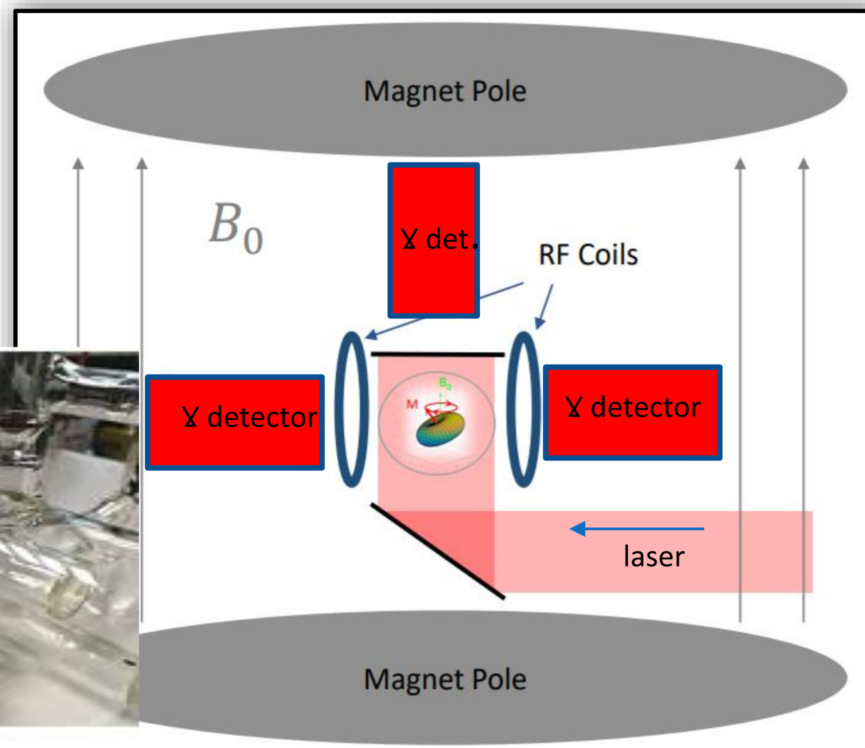
- Work on feasibility of the technique and improve on the published results:
  - Use long-lived Xe isomeric states
  - Optimise rf pulse sequence
  - Maintain polarization
  - Use state-of-the-art gamma-detectors
- => lower dose required to record signals
- Work on proof-of-principle experiment with commercial MRI scanner





gamma-MRI test setup

- Project:
  - Proof-of-principle gamma-MRI study
- Status:
  - 133mXe well produced at ISOLDE
  - Compact setup to polarize and detect asymmetry tested
- Next step: polarization of 133mXe



Summary and outlook

- Spin polarization seen in spatial distribution of nuclear decays:
- Beta decay asymmetry => Vud matrix element
- Beta-gamma-particle correlations => spin and parity of excited states
- Beta-NMR now applied to liquid biological samples, and not only
- Gamma-MRI: potentially new modality in medical imaging
- More applications possible, e.g. in nuclear and solid-state physics

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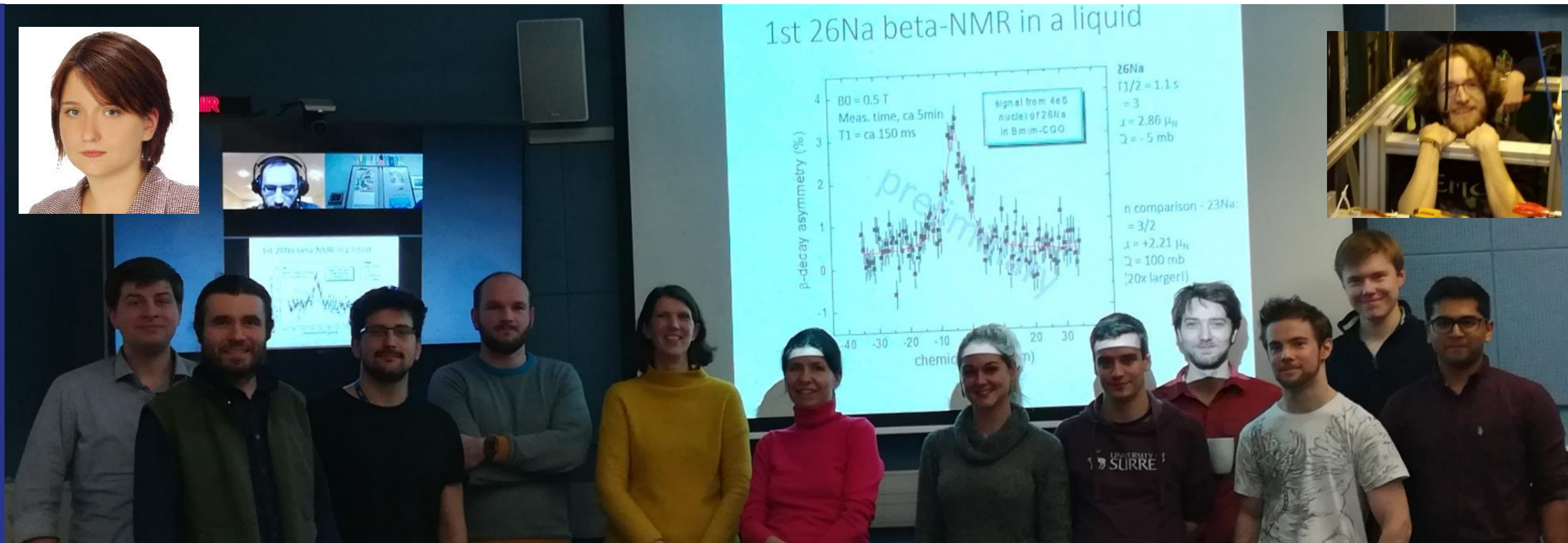

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