



**UNIVERSITÉ
DE GENÈVE**



**Spin-polarized exotic nuclei:
from fundamental interactions,
via nuclear structure,
to biology
and medicine**

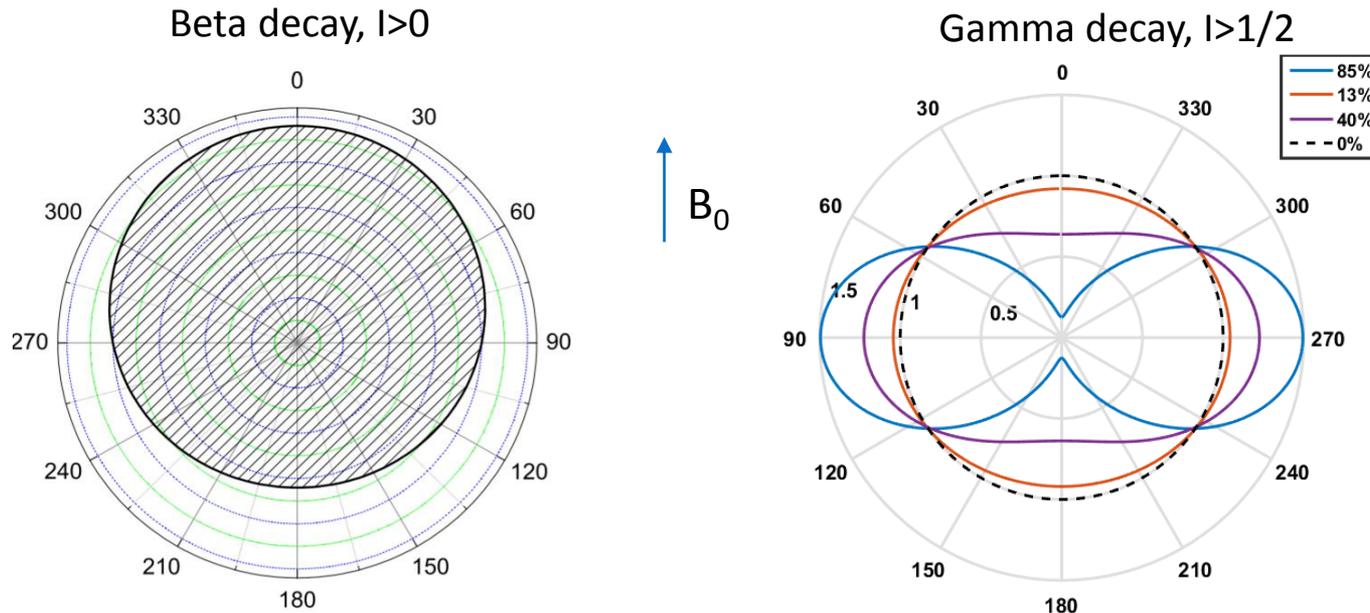
Magdalena Kowalska
UNIGE and CERN

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



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

$$a_1 = P a_\beta$$

$$W(\theta_r) = a_0 + a_2 \cos(2\theta_r) + a_4 \cos(4\theta_r) + \dots$$

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

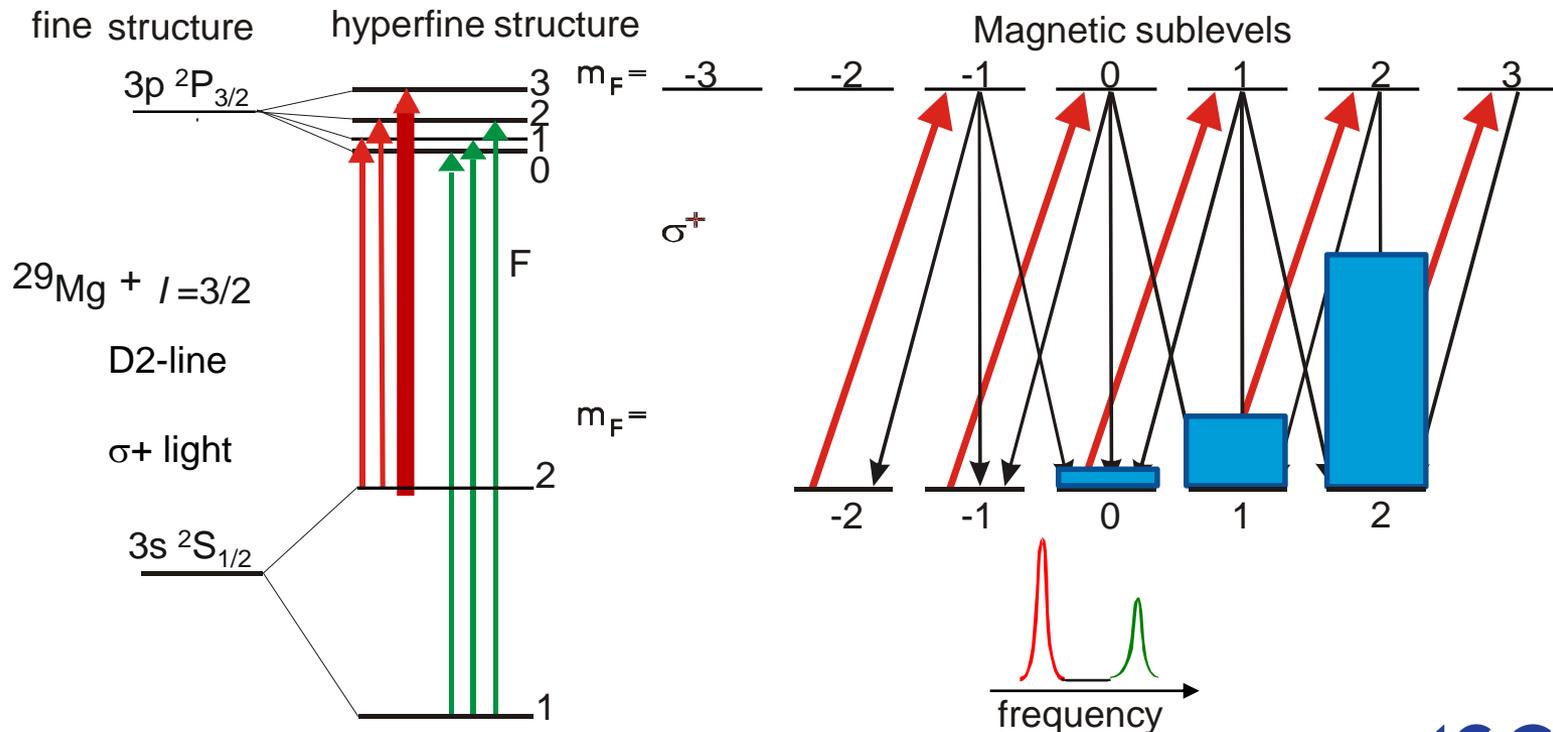
- Observed decay asymmetry can be used to:

- Probe underlying decay mechanism
- Determine properties of involved nuclear states
- Derive differences in nuclear energy levels³

$$\left\{ \begin{array}{ll} 1 & \text{for } I_f - I_i = -1 \\ \frac{1}{I_i + 1} \text{ (GT)} & \text{for } I_f - I_i = 0 \\ -\frac{I_i}{I_i + 1} & \text{for } I_f - I_i = 1 \end{array} \right.$$

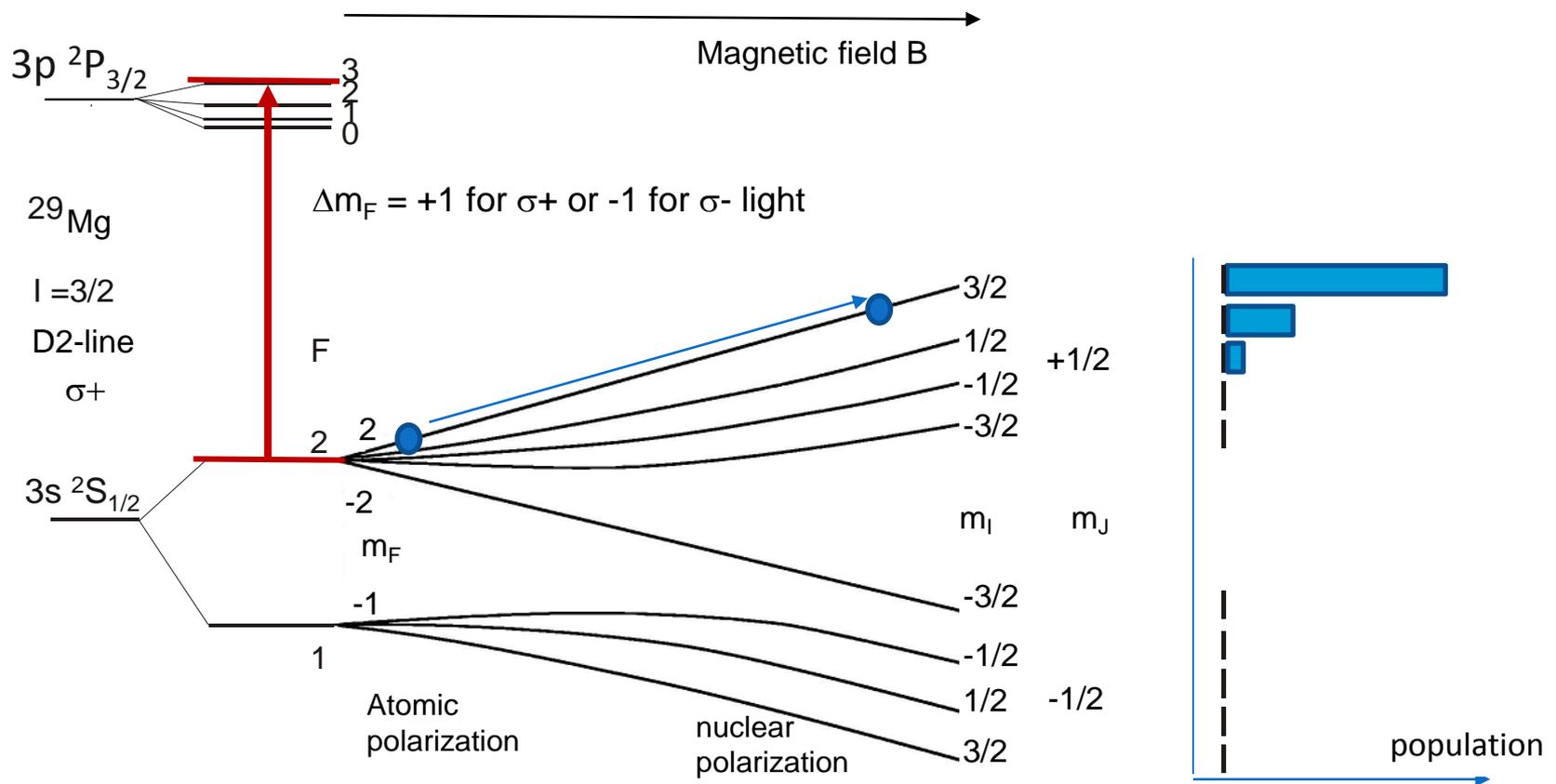
Spin polarization via optical pumping

- 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** ($P_F \rightarrow P_I$)
- Decoupling of electron (J) and nuclear (I) spins in strong magnetic field
- Observation of nuclear spin polarization P_I possible



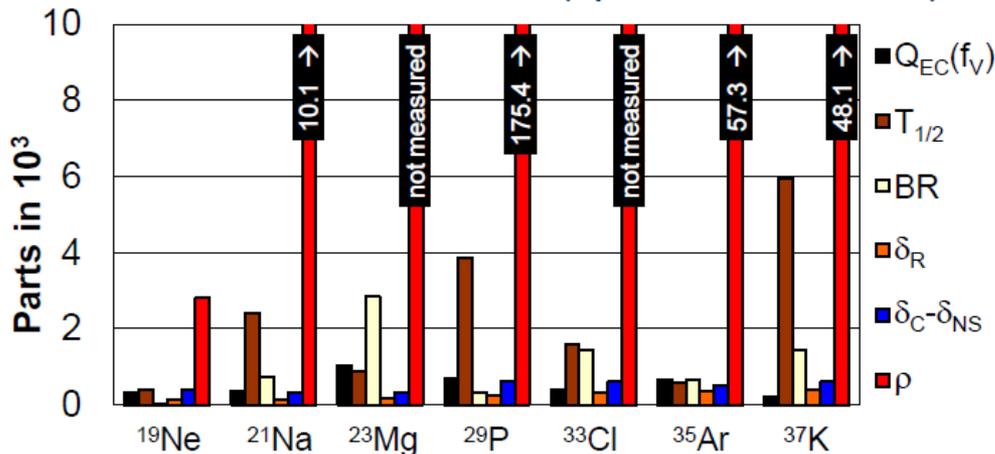
Decay asymmetry

Fundamental physics: measure V_{ud}

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



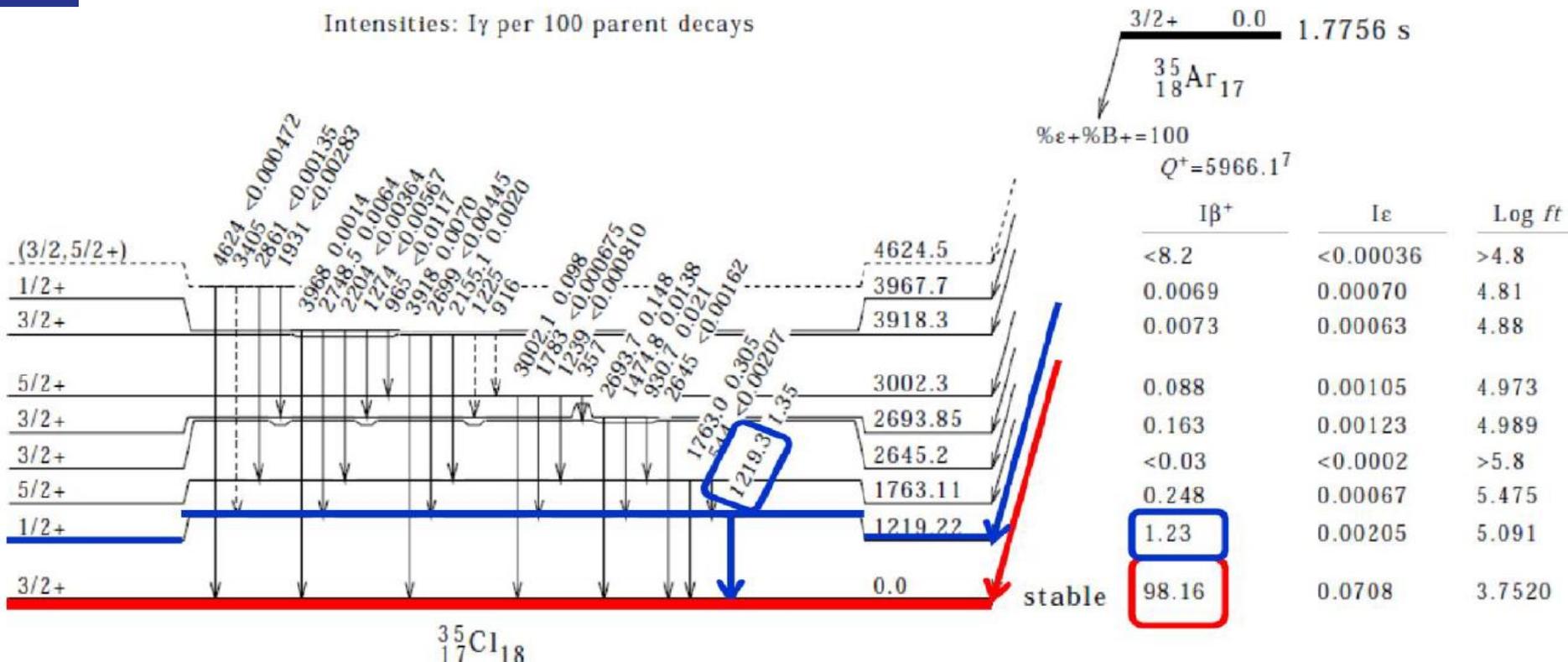
$$|V_{ud}| = 0.9719(17)$$

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

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

V_{ud} from $^{35}\text{Ar} \rightarrow ^{35}\text{Cl}$ mirror decay

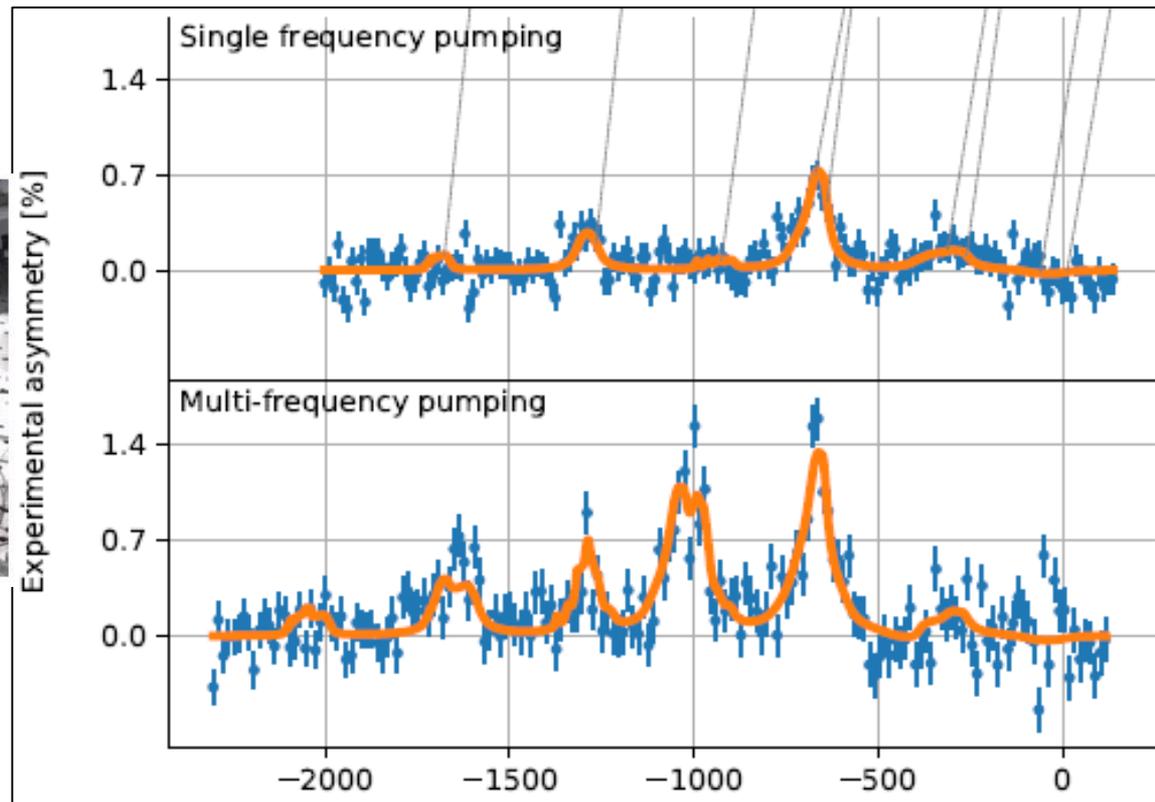
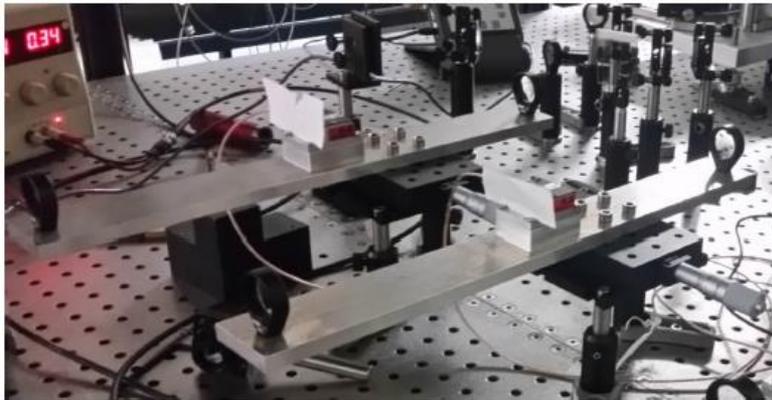
- Measurement of ^{35}Ar beta asymmetry factor a_β with 0.5% precision
 \Rightarrow Single V_{ud} value more precise than present weighed value



$a_\beta(\text{gs}) / a_\beta(\text{ex}) = \text{asymmetry}(\text{gs}) / \text{asymmetry}(\text{ex})$ and reference $a_\beta(\text{ex}) = 1$
 $\Rightarrow a_\beta(\text{gs}) = \text{asymmetry}(\text{gs}) / \text{asymmetry}(\text{ex})$ [both measured with our setup at ISOLDE]

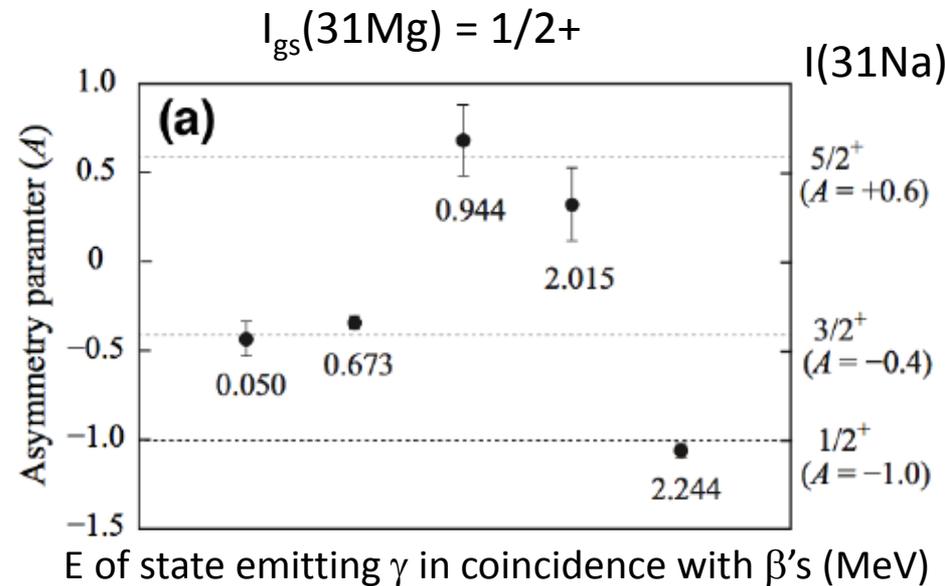
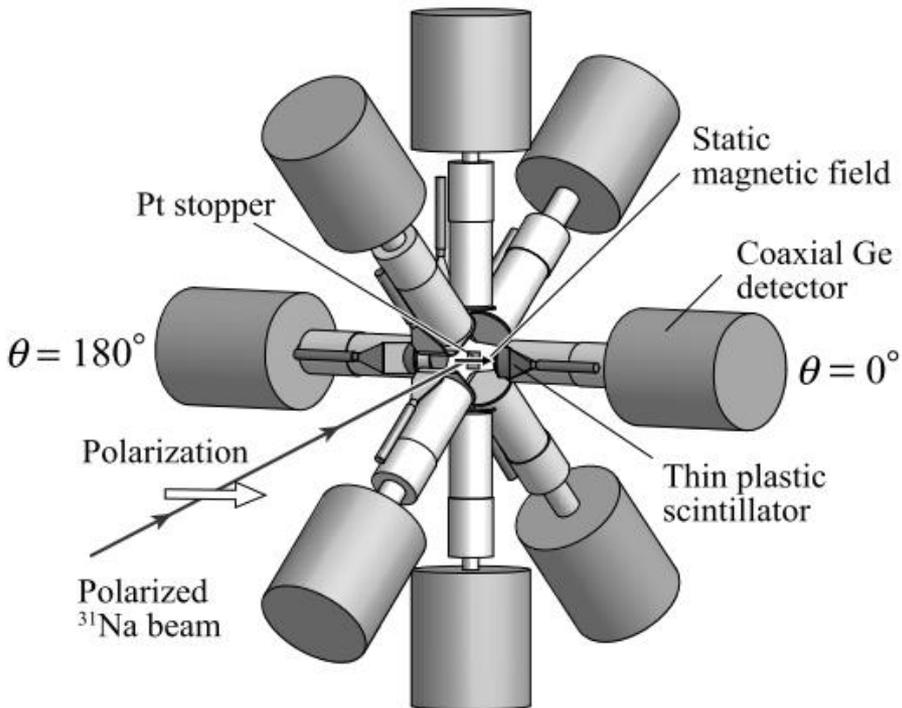
V_{ud} from ^{35}Ar decay: status

- Laser-polarization of ^{35}Ar 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)



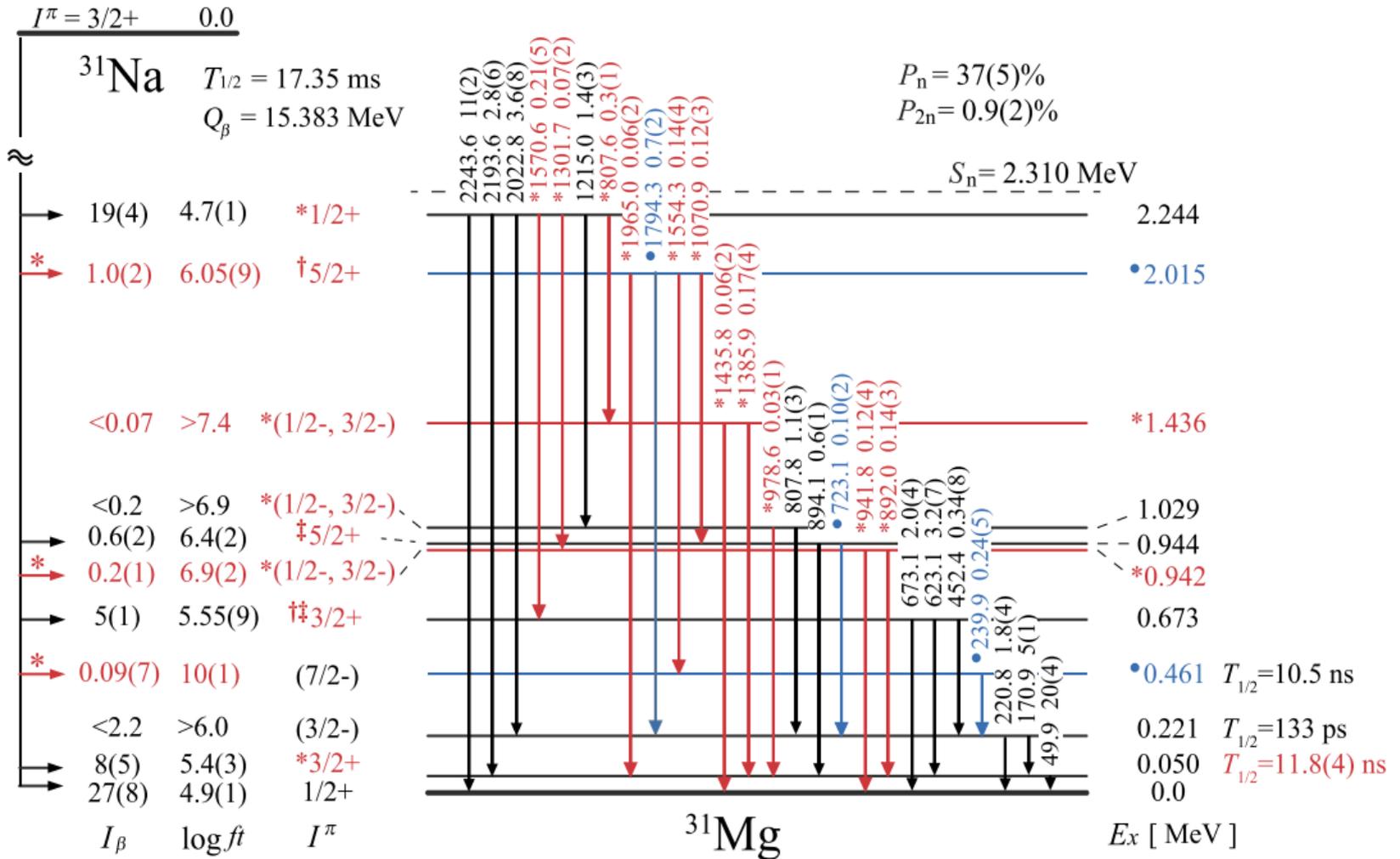
Spins/parities of states: β - γ correlations

- 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 $a_\beta = 1$ 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. $^{31}\text{Na} \rightarrow ^{31}\text{Mg}$:



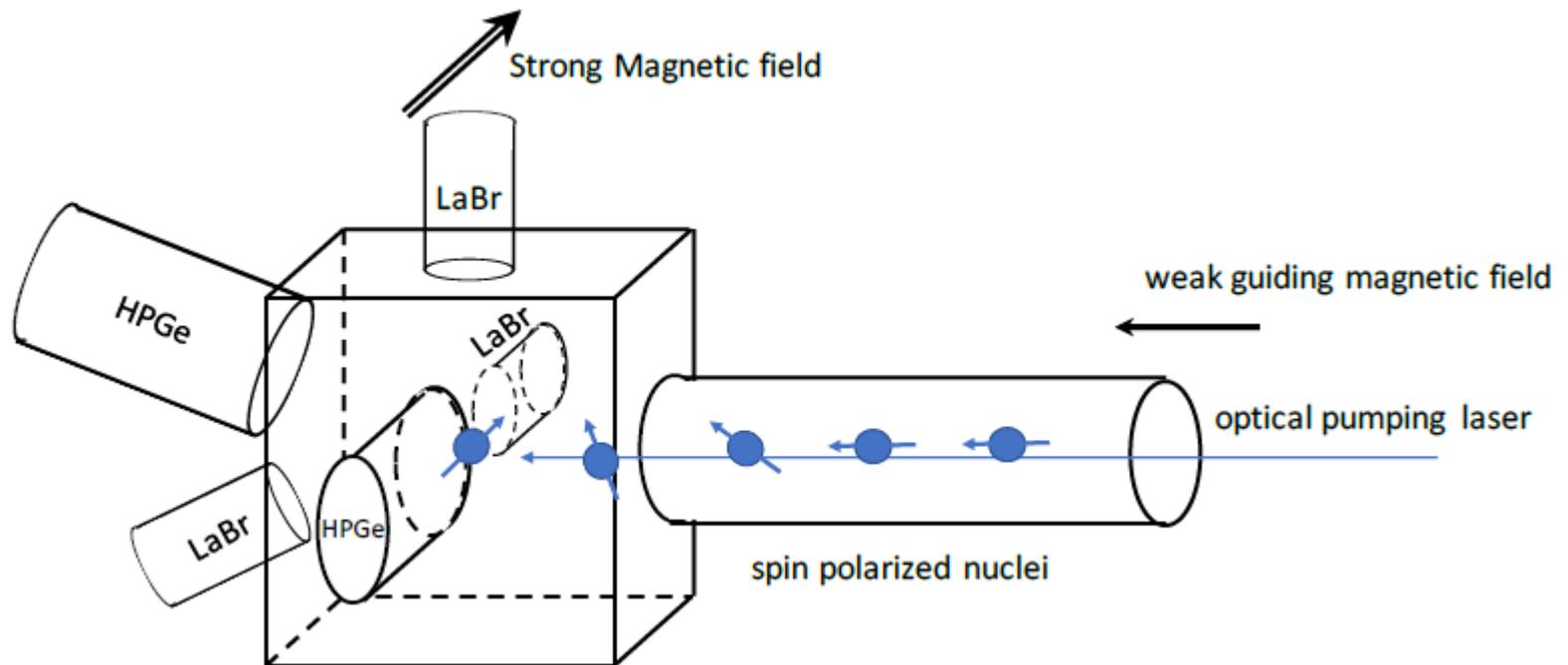
Spins/parities of states: β - γ correlations

Osaka setup at TRIUMF, e.g. $^{31}\text{Na} \rightarrow ^{31}\text{Mg}$



Spins/parities with our setup

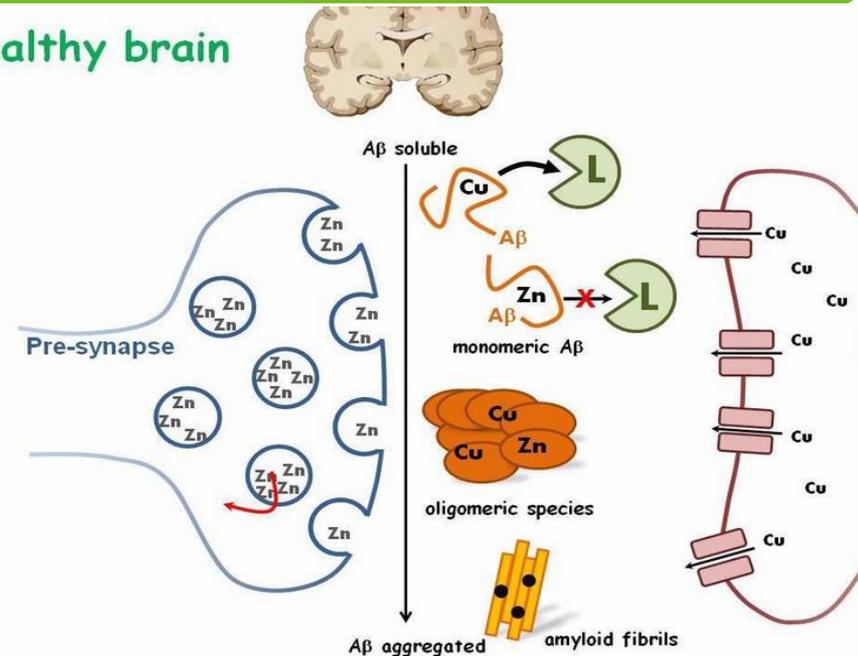
- Collaboration with Miguel Madurga, U Tennessee
- Letter of Intent submitted to ISOLDE (full proposal requested)
- To start: design of compact permanent magnets with smoothly
- Then: design of detector arrangement and their support
- Plan: setup ready when p's are back at CERN in 2021
- Later: include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



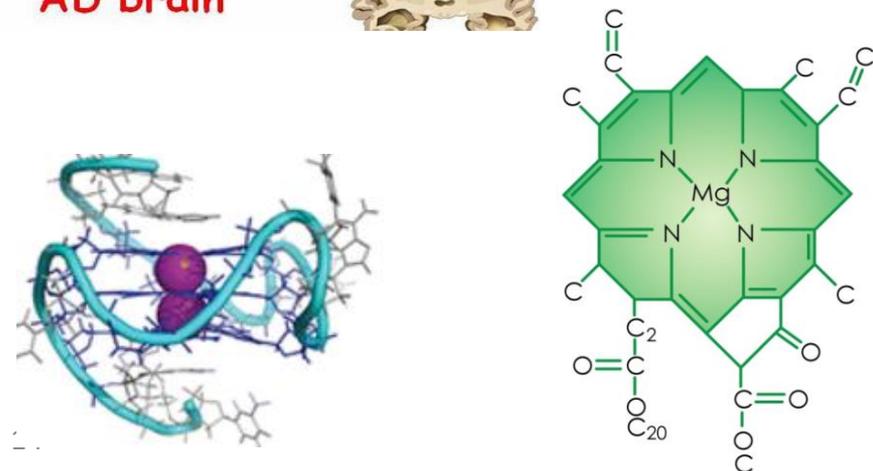
Nuclear Magnetic Resonance

- 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

healthy brain



AD brain



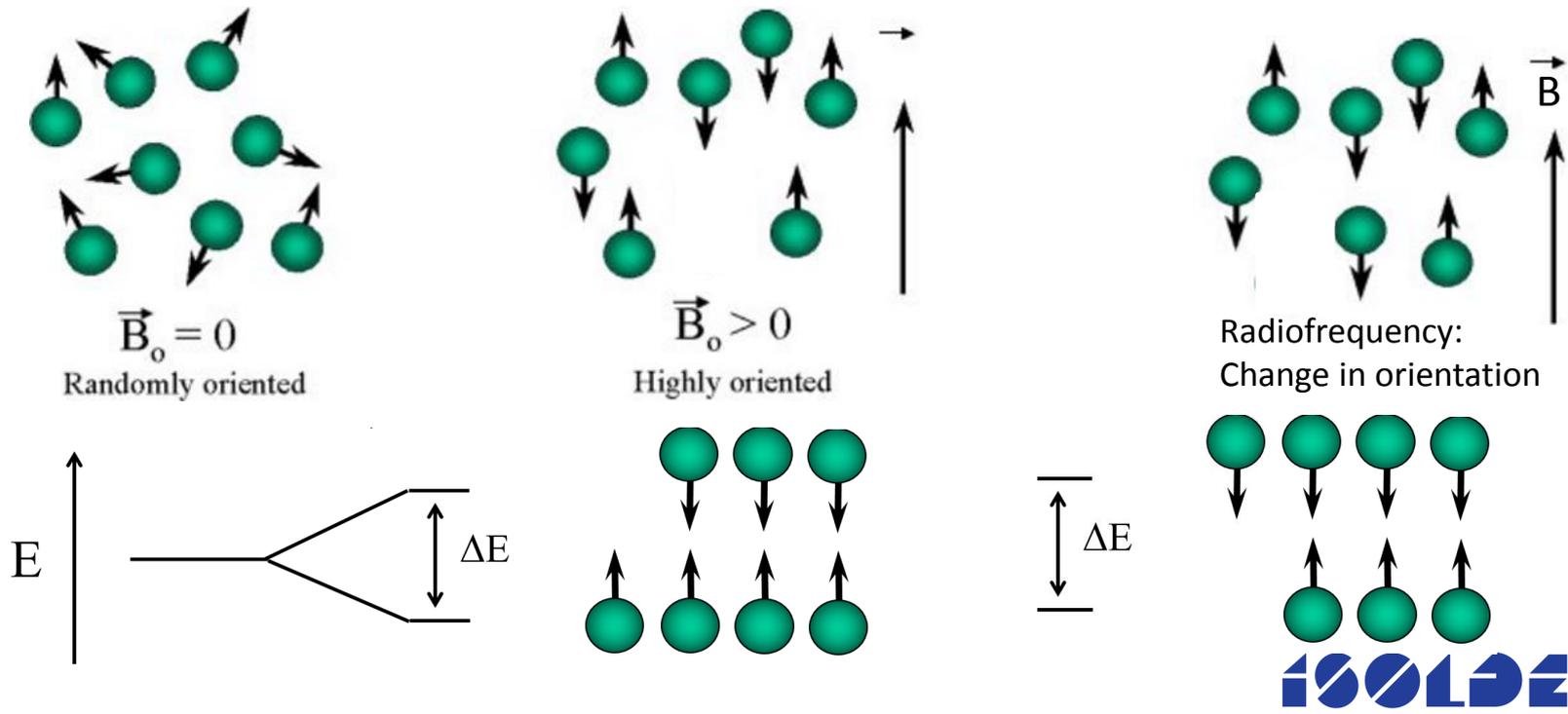
NMR principles

Participants:

- Probe nuclei with spin different from 0
- Sample/ environment

Magnetic field

- Strong static field (B_0) -> when different at different positions -> **MRI (Magnetic Resonance Imaging)**
- Weaker field (B_1) oscillating at radio-frequency (MHz)



NMR in nuclear physics

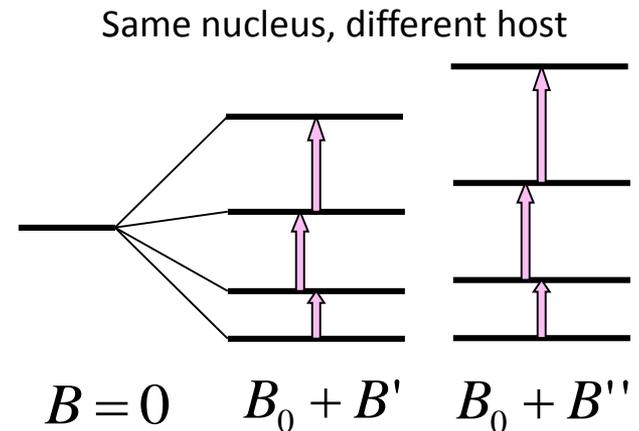
Method to determine precisely magnetic & quadrupole moments of short-lived nuclei

- **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



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

● Example: spin and magnetic moment of ^{31}Mg

- M. Kowalska, PhD Thesis, U Mainz 2006 ¹⁶

NMR in (chemistry and) biology

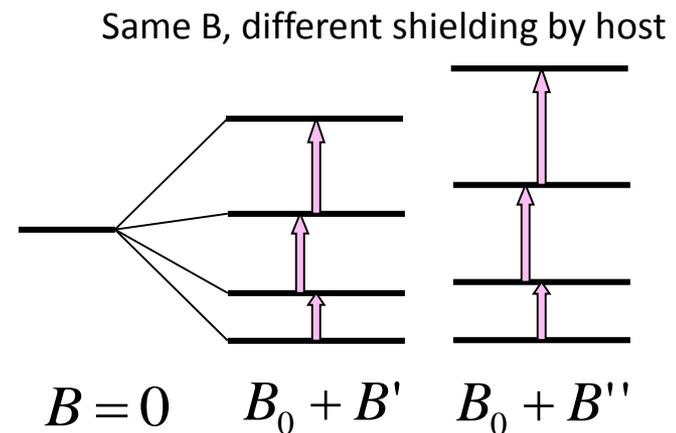
Most versatile method to study structure and dynamics of molecules in solution

- **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

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

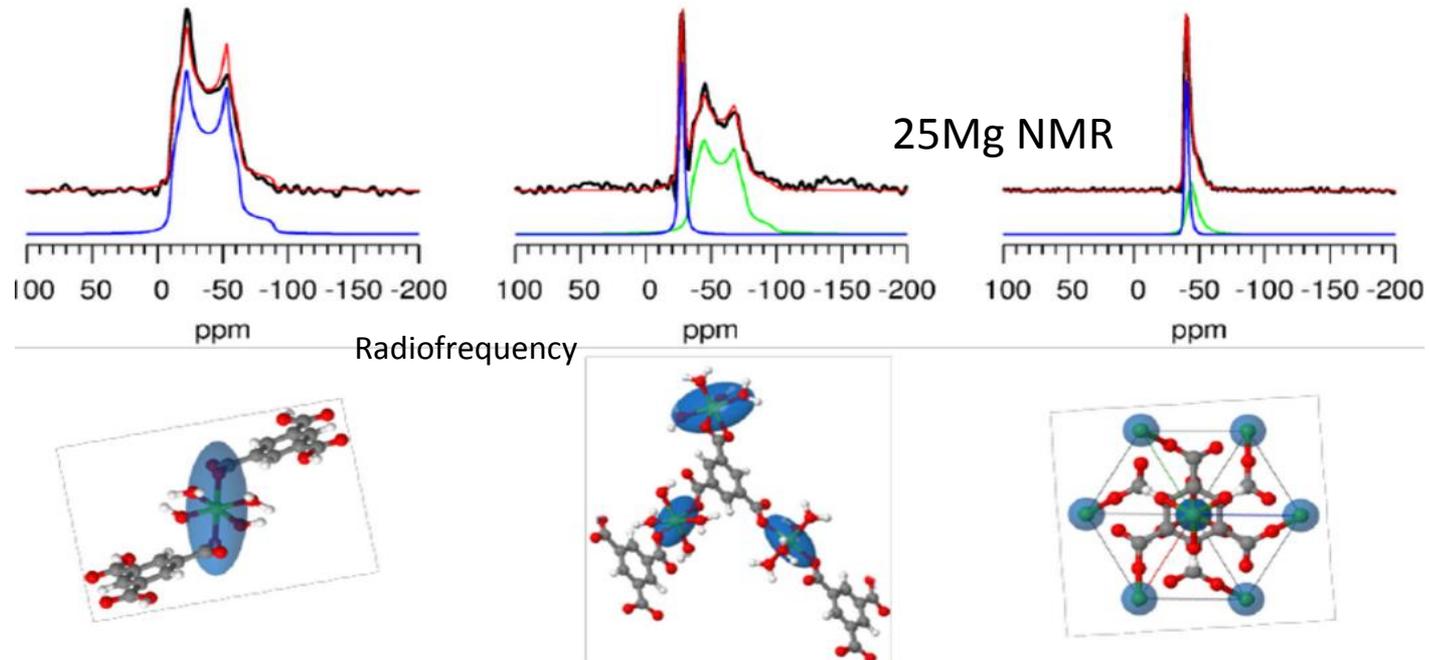
known



- **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

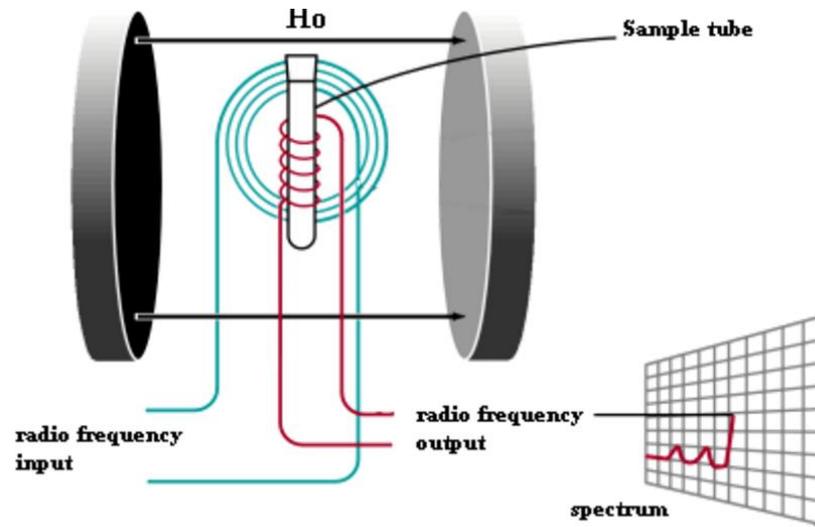
- NMR can provide info on location & evolution of metal-ion binding to biomolecules
 - NMR on ^{23}Na , ^{25}Mg , ^{63}Cu , ^{67}Zn bound to the biomolecule



- 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

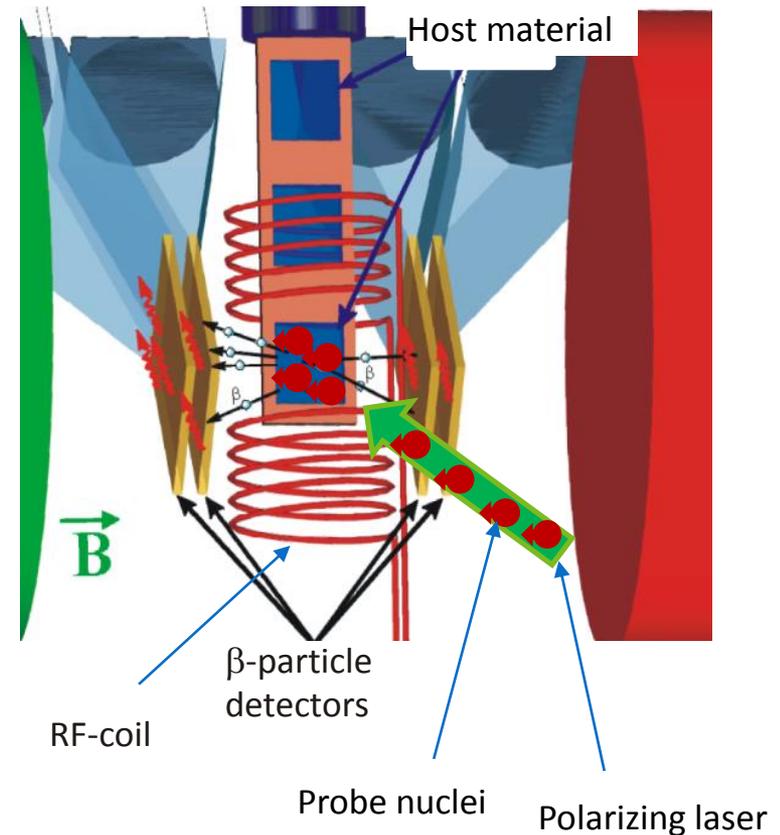
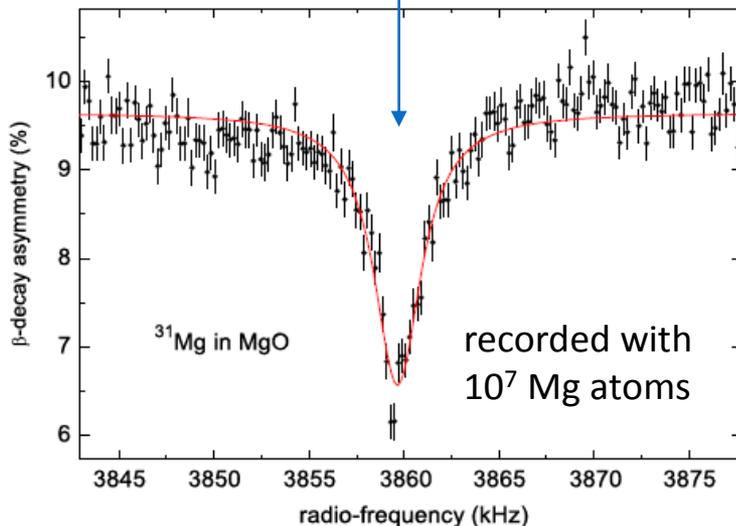
NMR limitation: sensitivity

- 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



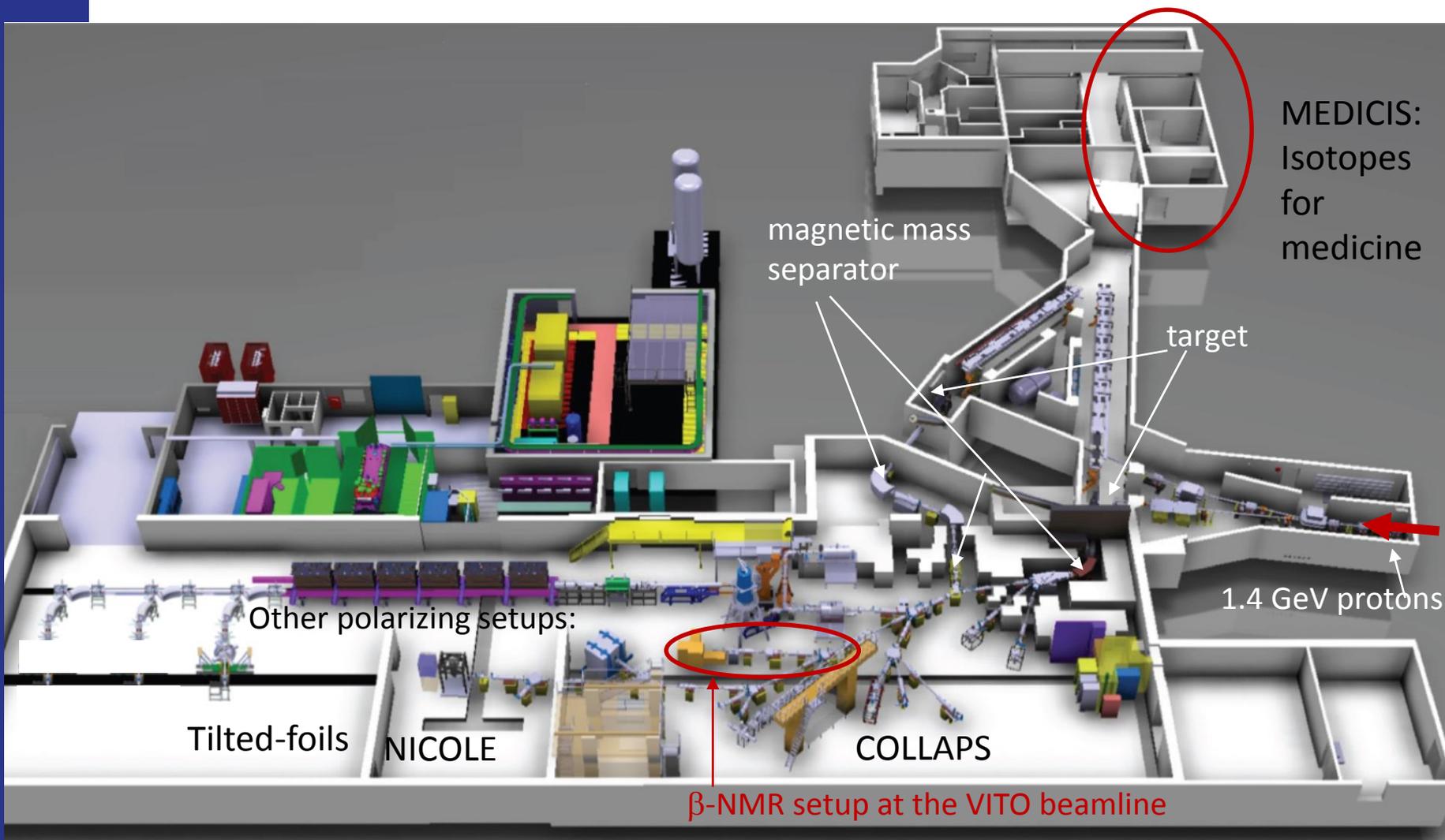
Beta(-detected) NMR

- 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**



Spin-polarization & β -NMR at ISOLDE-CERN

Laser-polarization and β -NMR at VITO beamline



ISOLDE selection of radio-nuclei

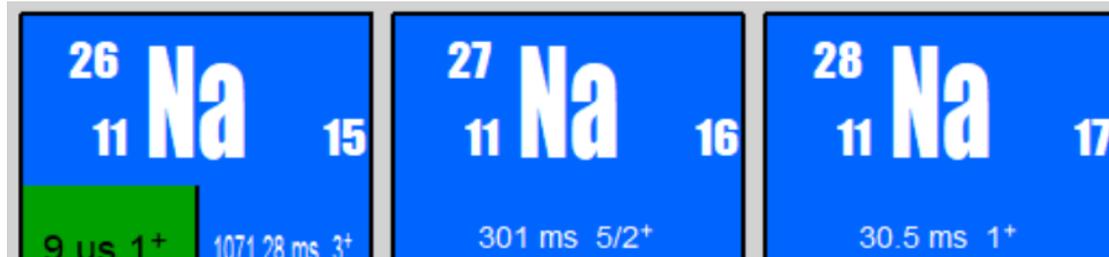
- Over 1200 isotopes from 80 chemical elements used for experiments

- Radioactive half-lives: >10 ms

- Laser-polarized metals:

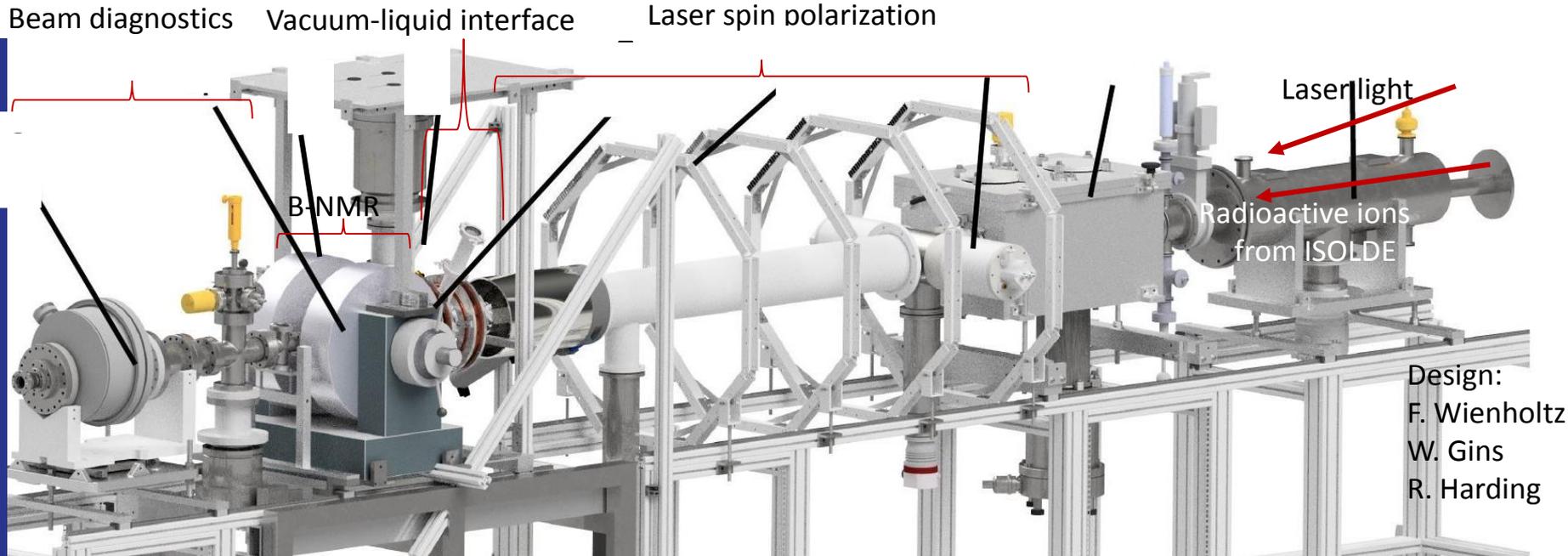
- Already polarized at ISOLDE

- Planned within this project



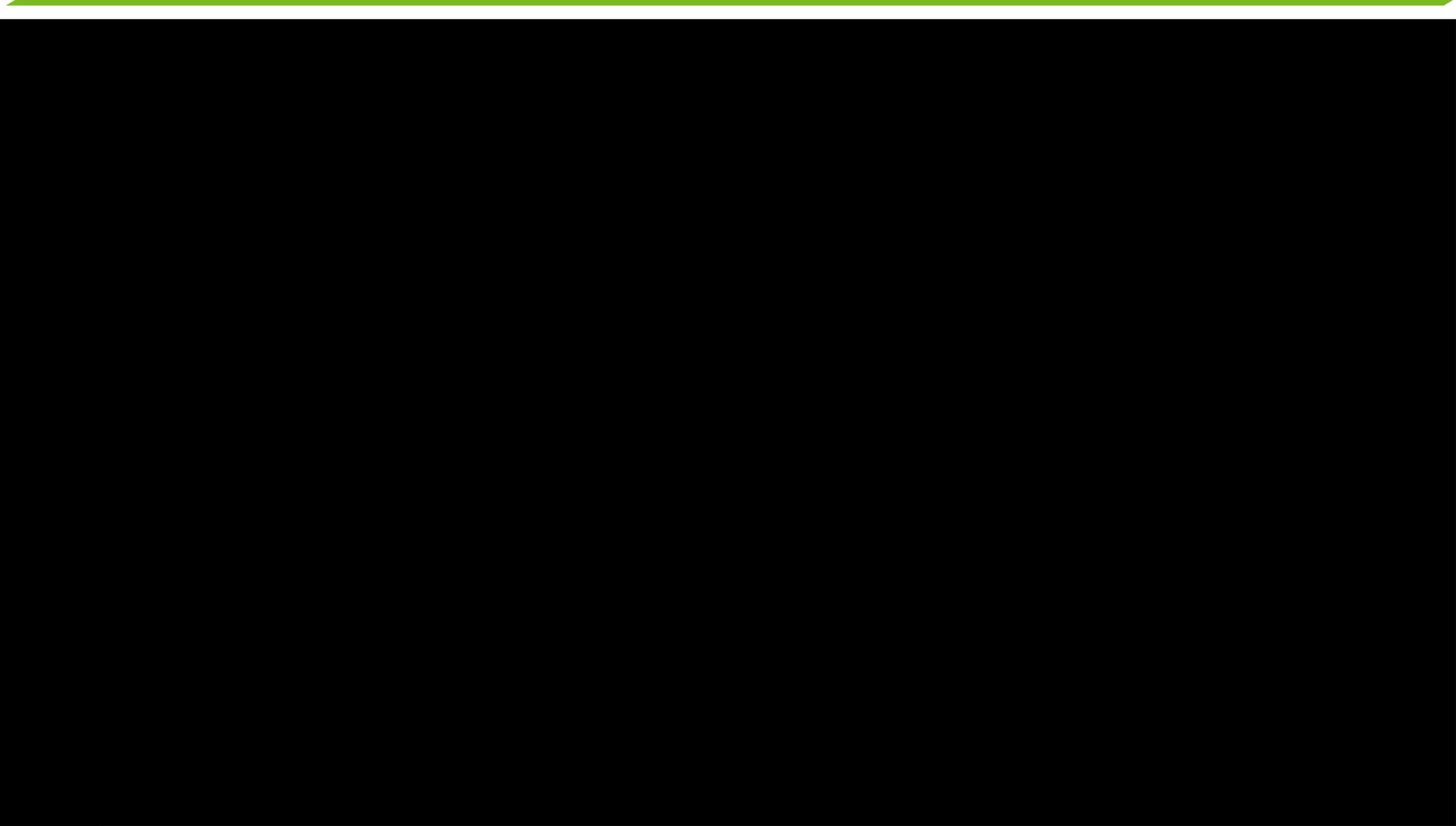
1	1 H																	2 He	
2	3 Li	4 Be											5 B	6 C	7 N	8 O	9 F	10 Ne	
3	11 Na	12 Mg											13 Al	14 Si	15 P	16 S	17 Cl	18 Ar	
4	19 K	20 Ca	21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn	31 Ga	32 Ge	33 As	34 Se	35 Br	36 Kr	
5	37 Rb	38 Sr	39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd	49 In	50 Sn	51 Sb	52 Te	53 I	54 Xe	
6	55 Cs	56 Ba	* 71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn	
7	87 Fr	88 Ra	** 103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Ds	111 Rg								
* Lanthanides		*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb			
** Actinides		**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No			

Experimental setup

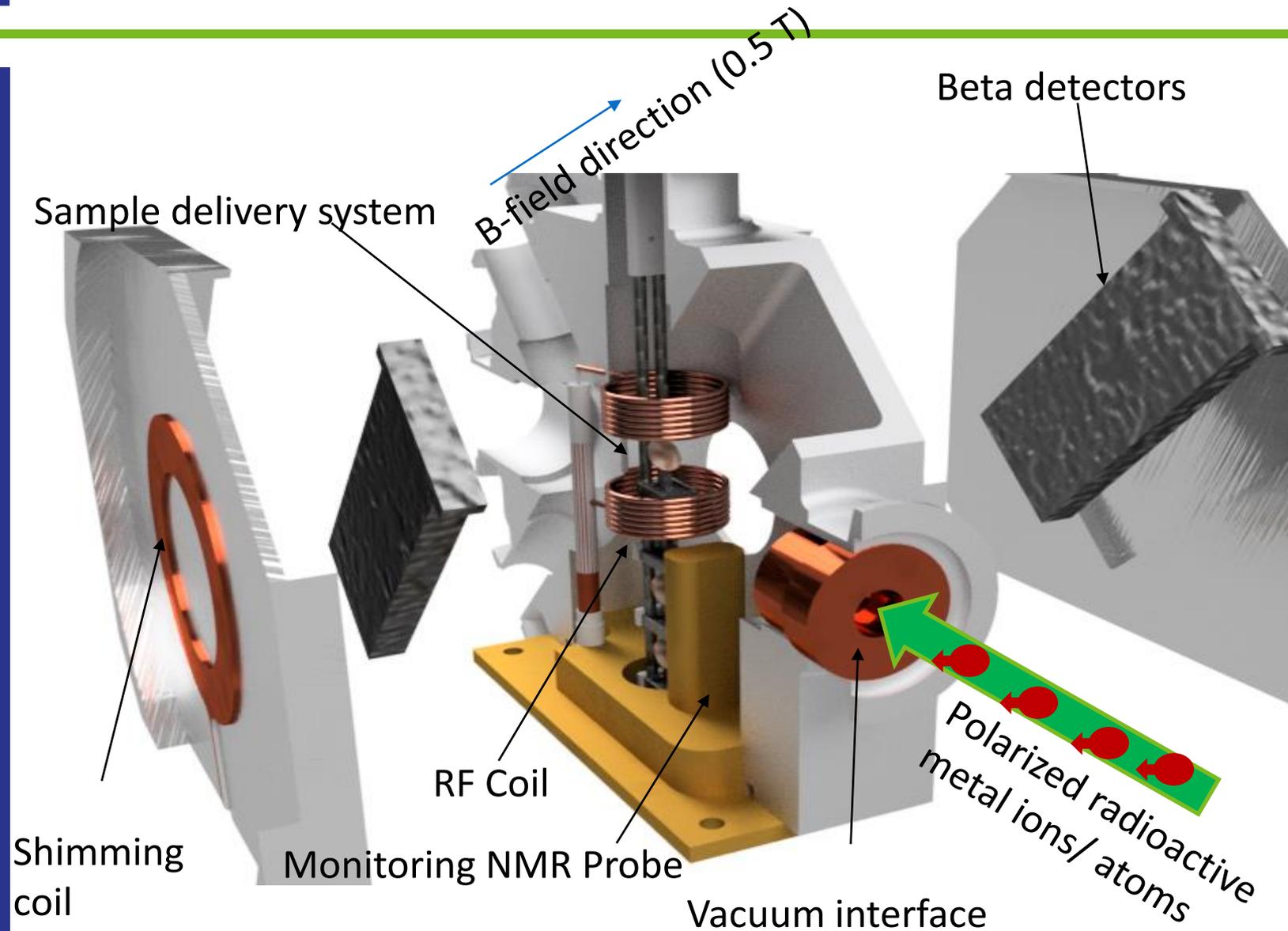


M. Kowalska et al., J. Phys. G: Nucl. Part. Phys. 44 (2017) 084005
W. Gins et al., submitted to Nucl. Instr. and Meth. A (2018)
W. Gins, PhD Thesis, KU Leuven (2019)

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



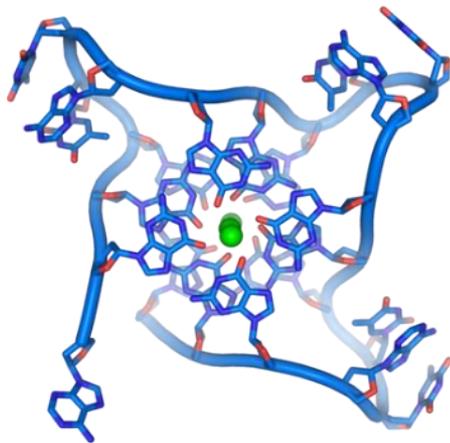
Bio-Beta-NMR chamber



First bio-study: Na⁺ & G-quadruplexes

DNA G-quadruplexes:

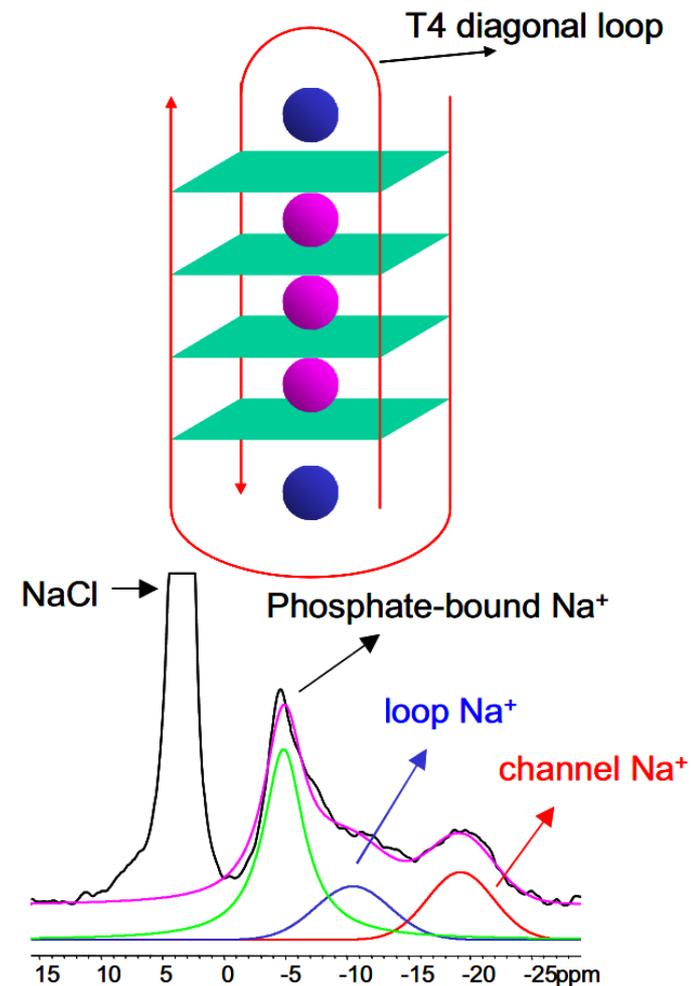
- Guanine-rich DNA fragments
- Found in nature, e.g. in telomeres or oncogenes
- Synthesised for novel applications
- Important in different diseases



Alkali metals in DNA G-quadruplexes

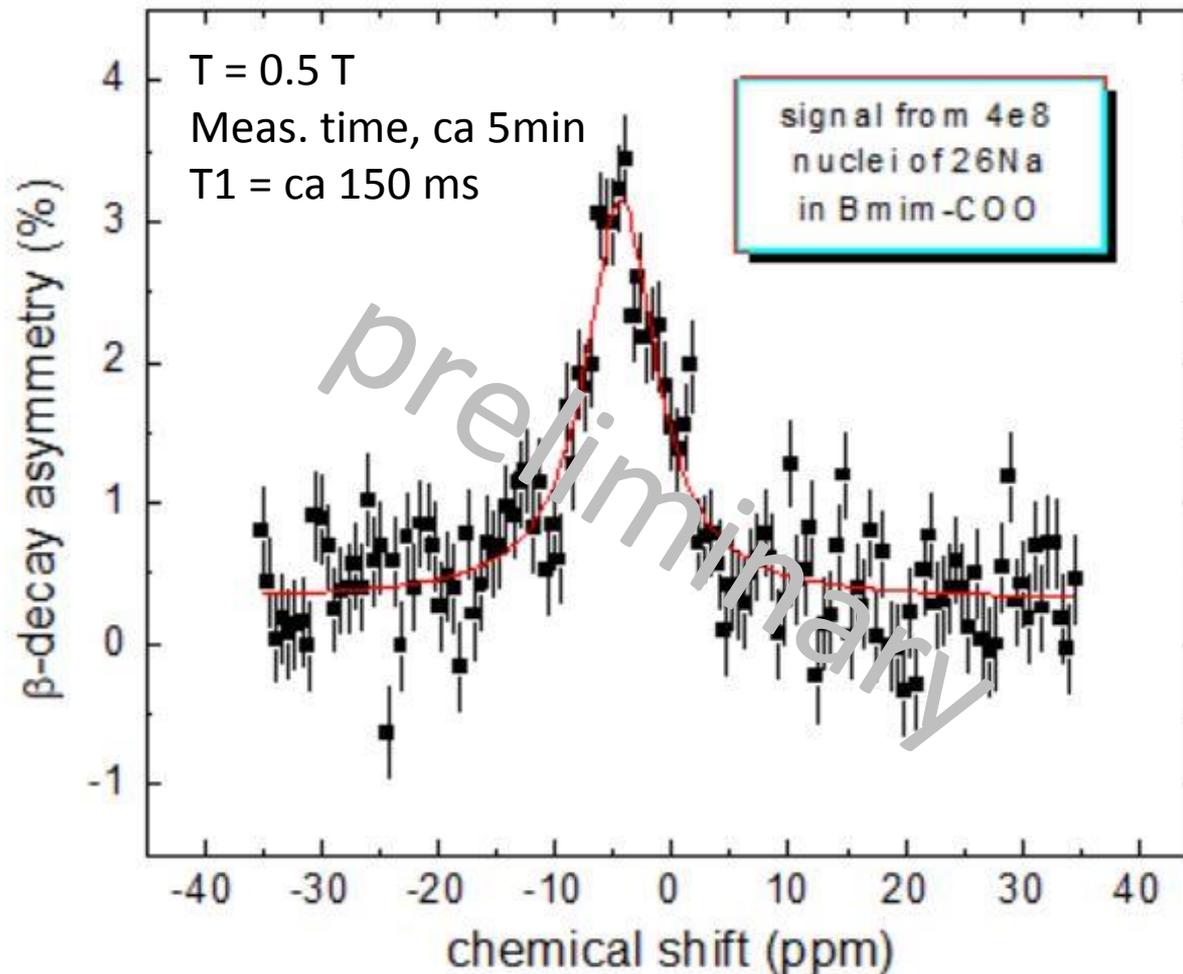
- Important for their formation, stability and structural polymorphism
- Until recently considered invisible in conventional Na⁺/K⁺ NMR

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



1st liquid beta-NMR results

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



^{26}Na

$T_{1/2} = 1.1 \text{ s}$

$I = 3$

$\mu = 2.86 \mu_N$

$Q = -5 \text{ mb}$

In comparison - ^{23}Na :

$I = 3/2$

$\mu = +2.21 \mu_N$

$Q = 100 \text{ mb}$

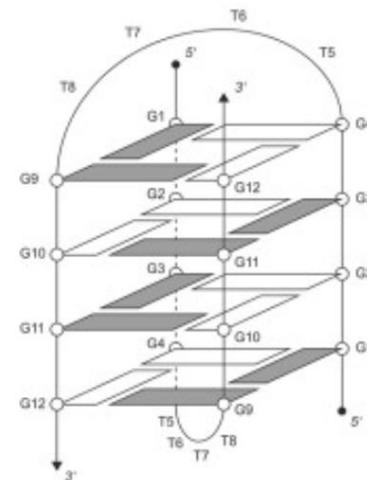
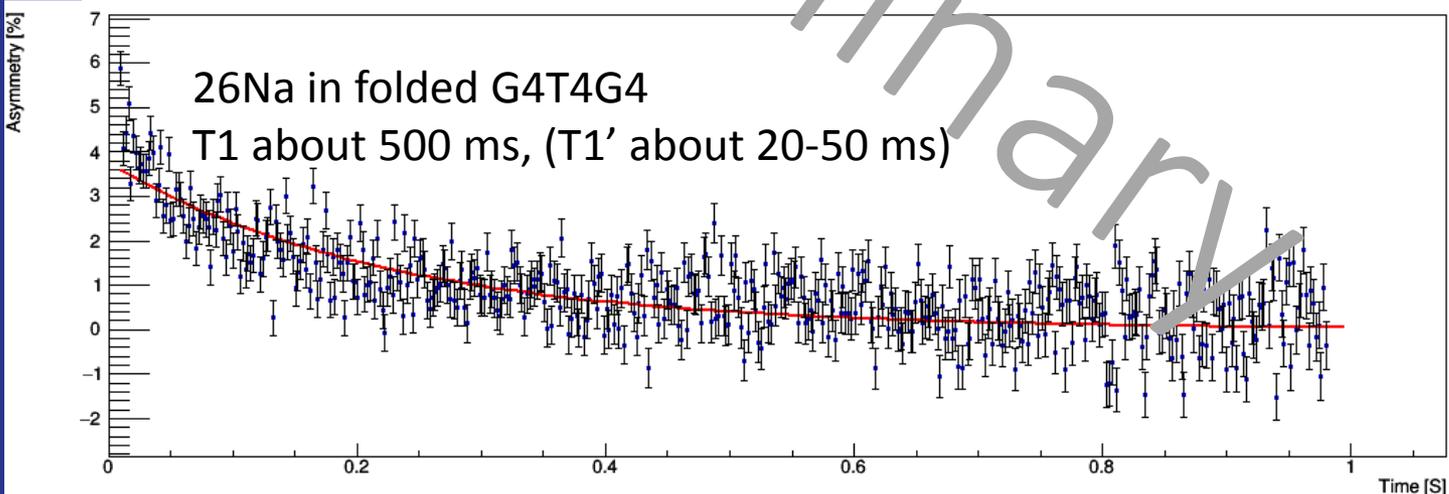
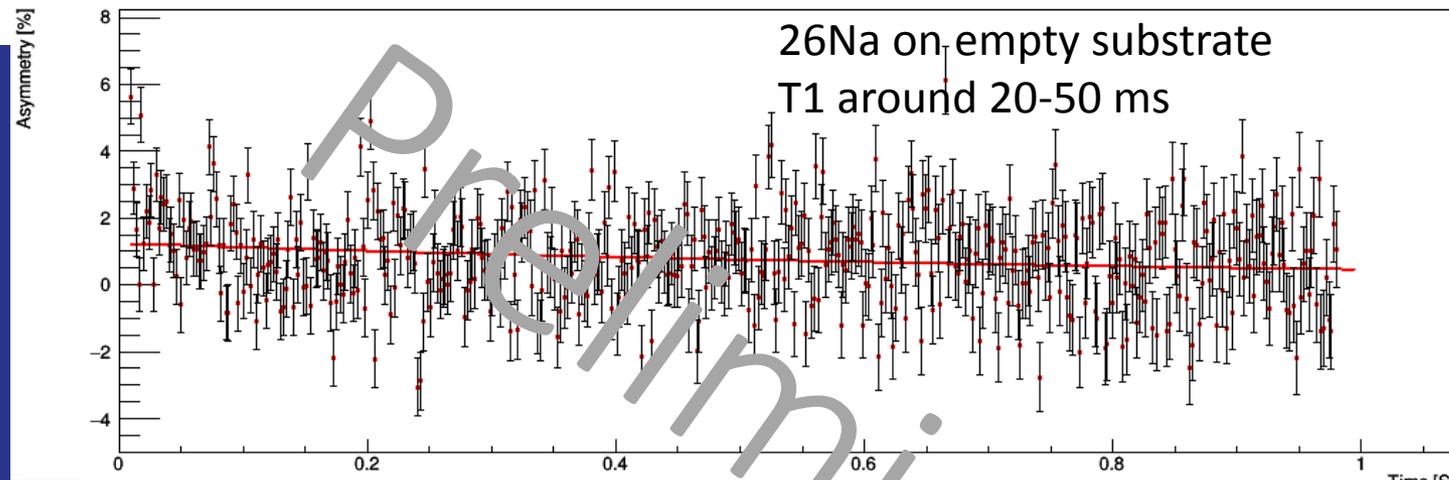
(20x larger!)

Resonance width will allow determining chemical shifts expected for Na

β -NMR: ^{26}Na T1 in G4T4G4 G-quadruplex

May 2018

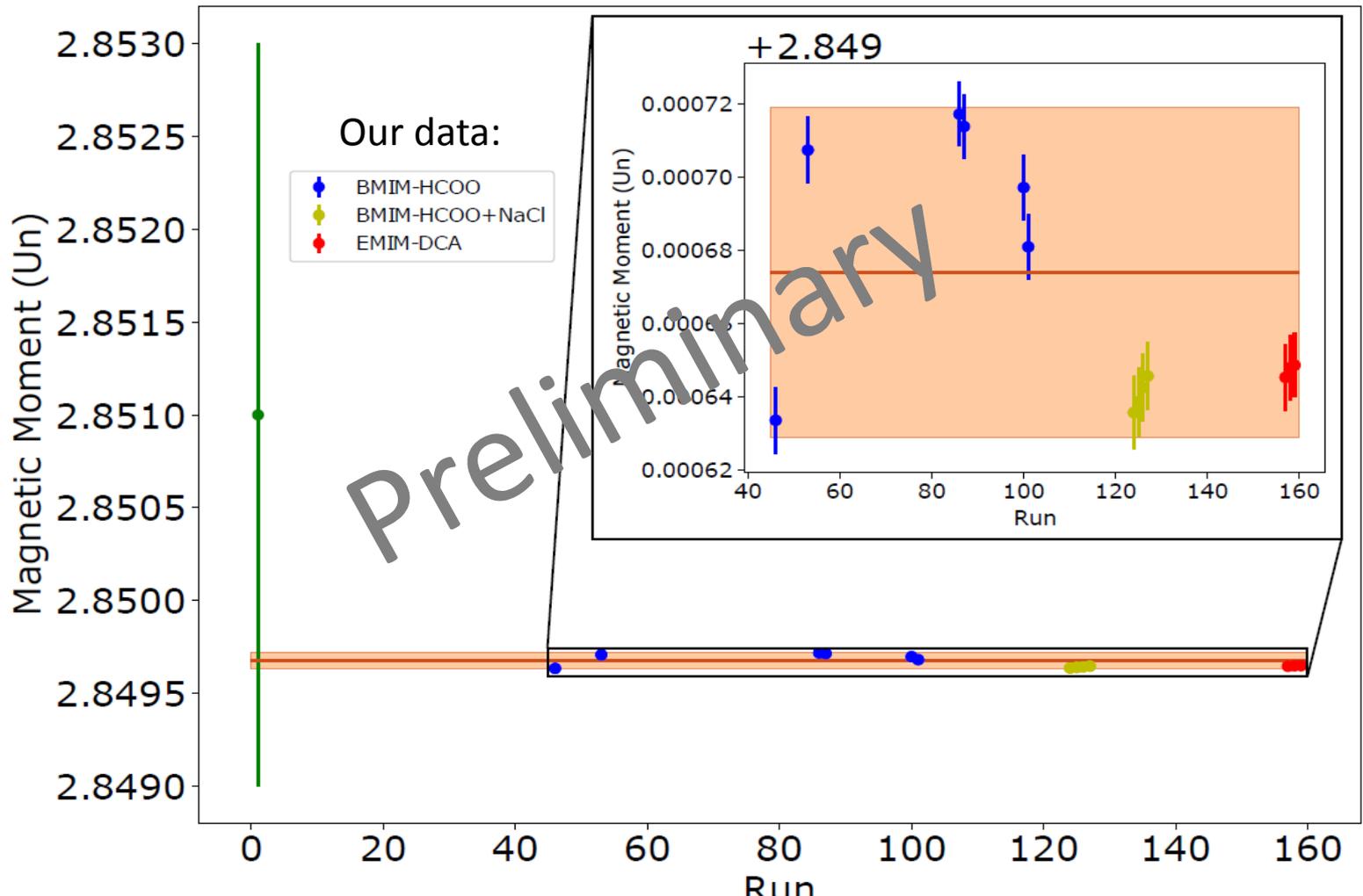
Folded
G4T4G4
DNA sequence
on substrate



- 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

Physics result: ultraprecise $\mu(26\text{Na})$

- (β -)NMR resonances in liquids are very narrow:
=> magnetic moment of nuclei can be determined with much higher precision



Beyond 2018

Already polarized at ISOLDE:

8,9Li

11Be

26-28Na

29,31Mg

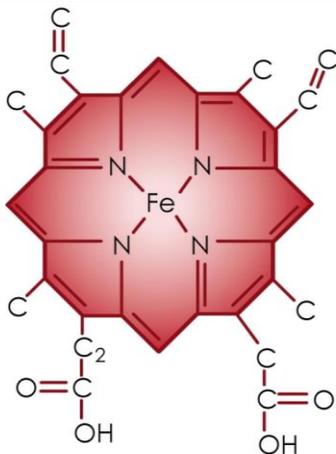
Feasible and planned soon:

37,49K

39,51Ca

58,74,75Cu

75,77Zn

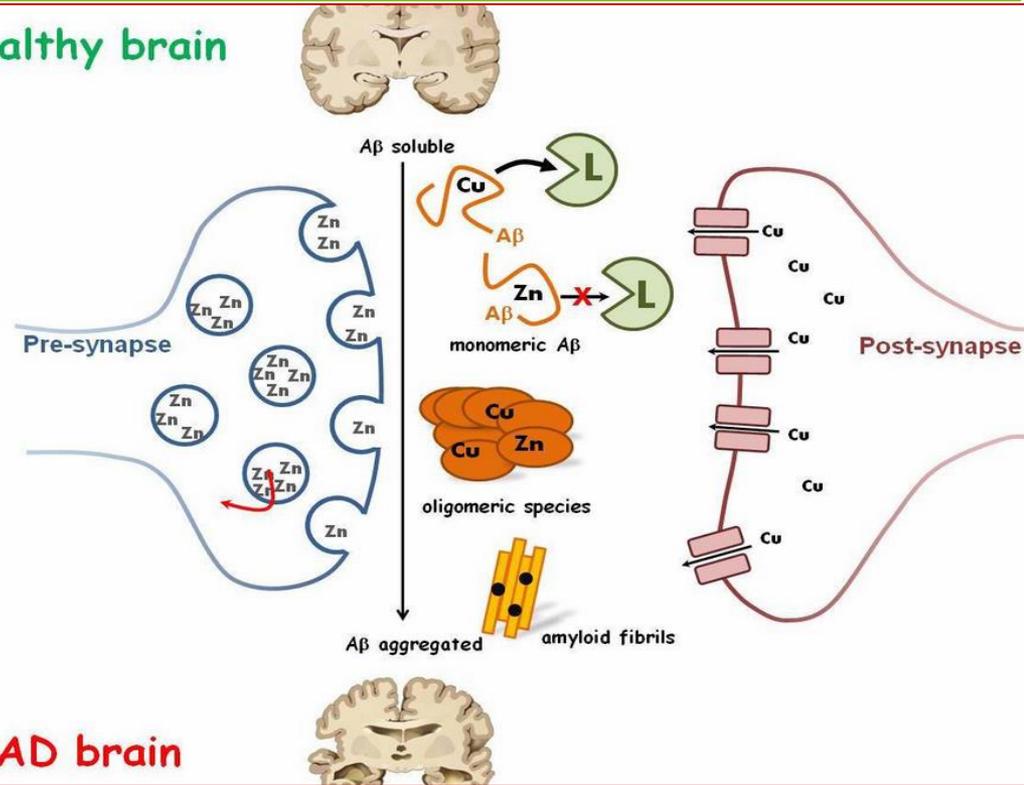


Human Blood Hemoglobin

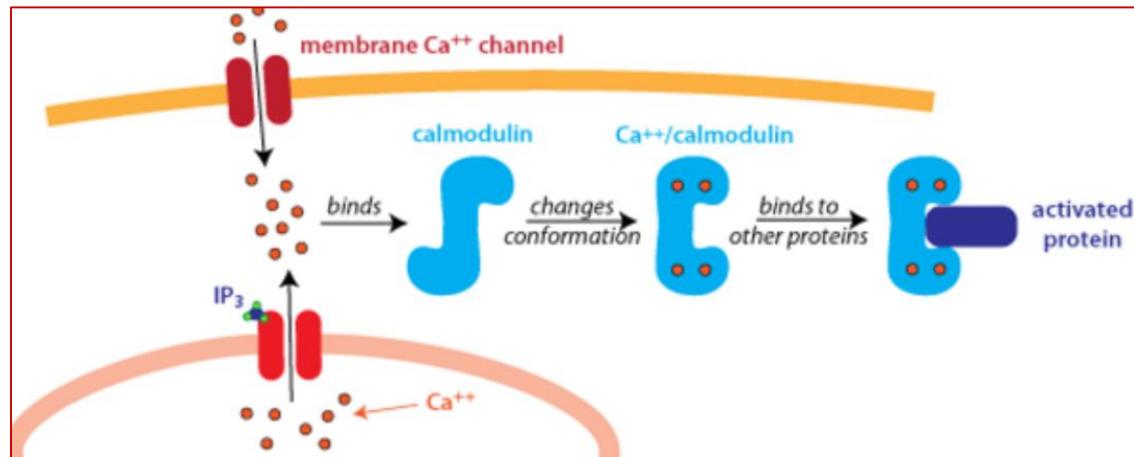


Plant Chlorophyll

healthy brain



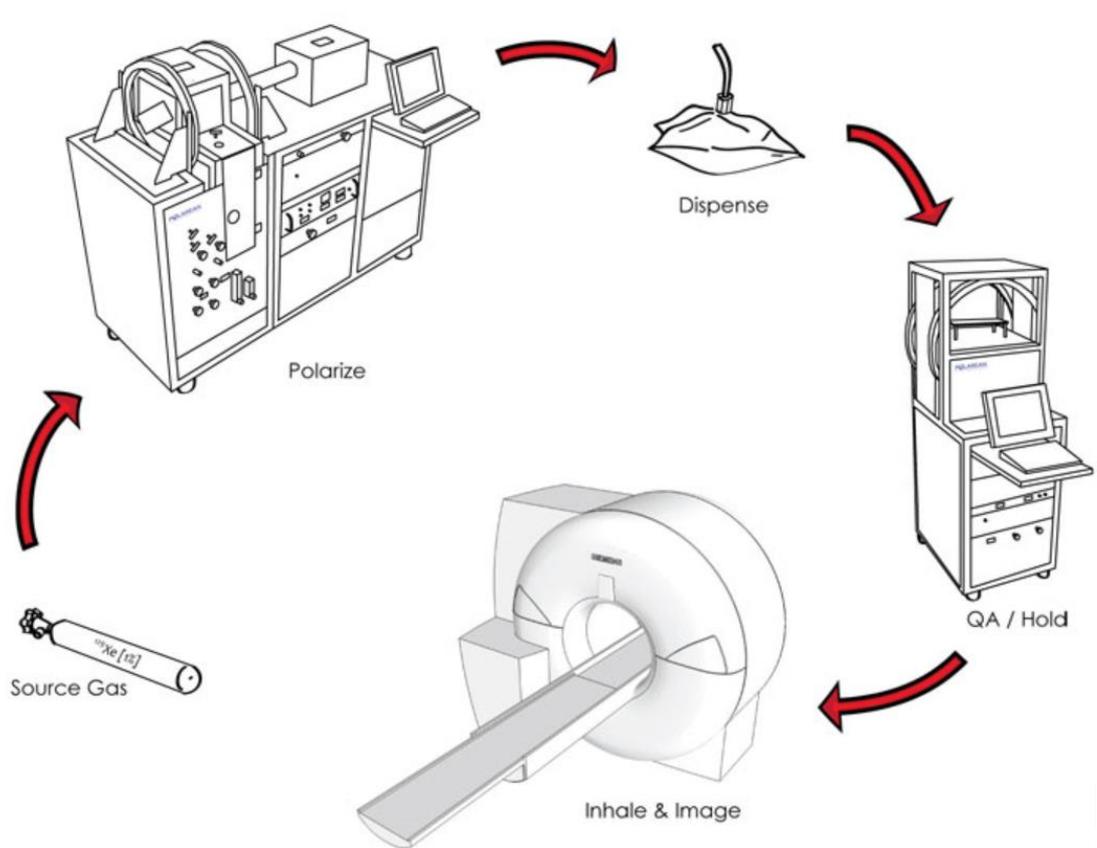
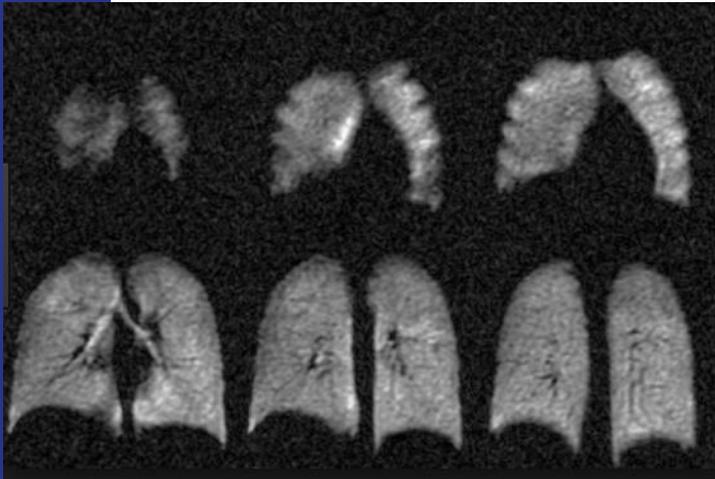
AD brain



Magnetic resonance imaging

MRI with hyperpolarized nuclei

- MRI: high resolution, but small sensitivity
- Hyperpolarization: increase in sensitivity by up to $1e5$
- Best example: ^{129}Xe :
 - 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:

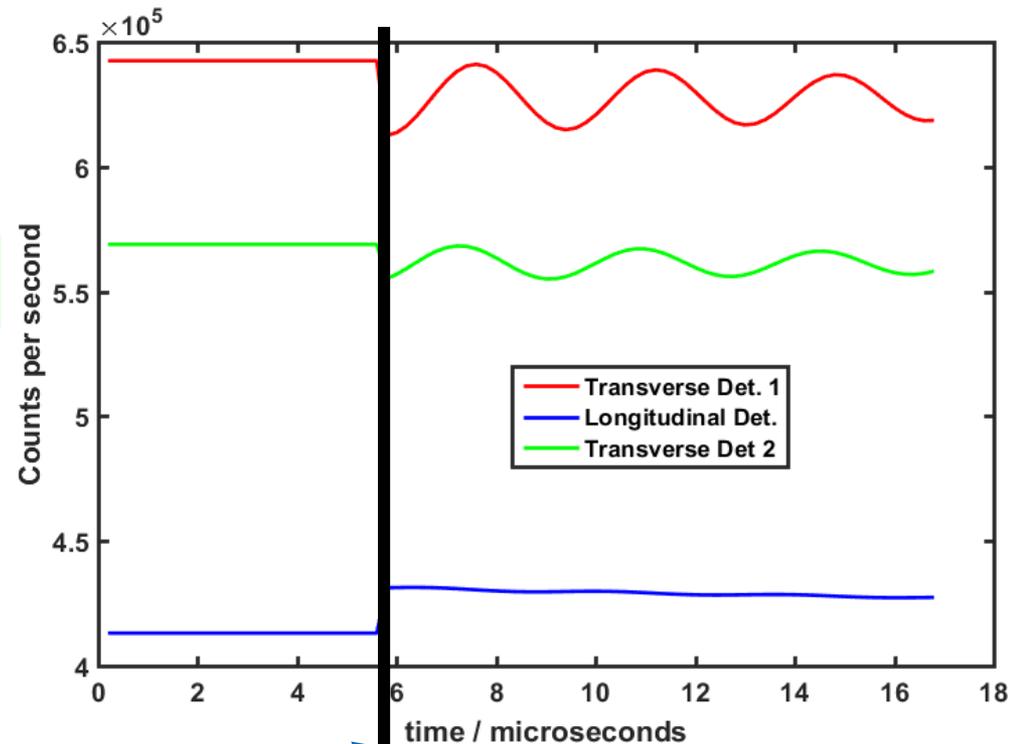
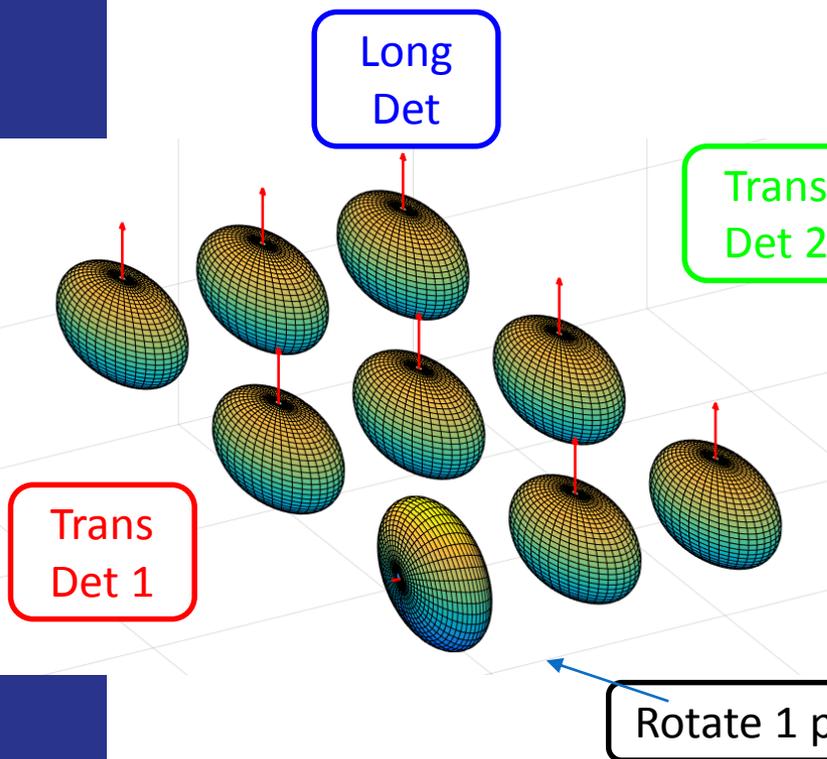
	Detection efficiency	Spatial resolution
PET and SPECT	High (PET>SPECT)	Low (e.g. >5mm for ^{82}Rb)
MRI	low	High

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

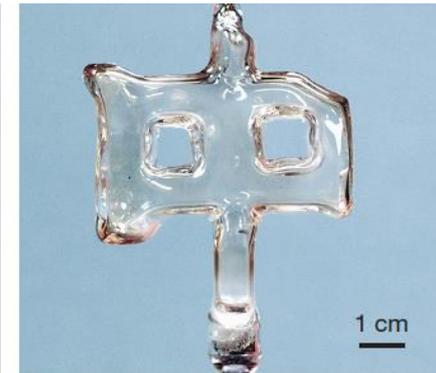
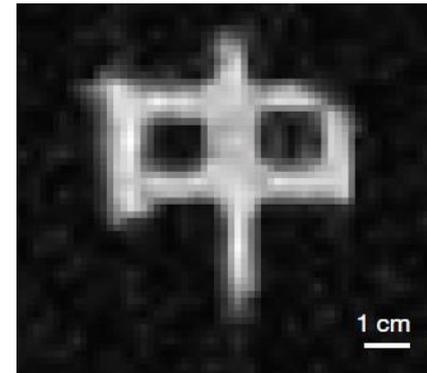
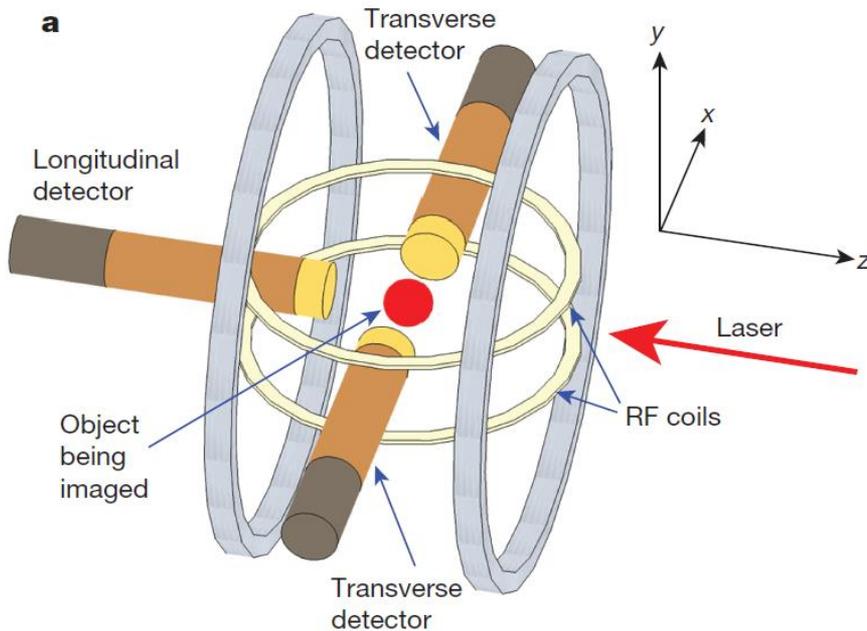
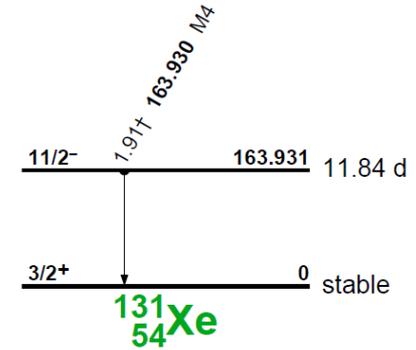
- **Voxel size**
 - defined by slope of B-field gradients and spectral width of rf pulse
 - more nuclei -> smaller pixels possible up to B gradient and rf limit
- **1 pixel in resonance:**
 - change in total gamma counts visible in each detector
 - Degree of change proportional to number of nuclei in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



First gamma-MRI

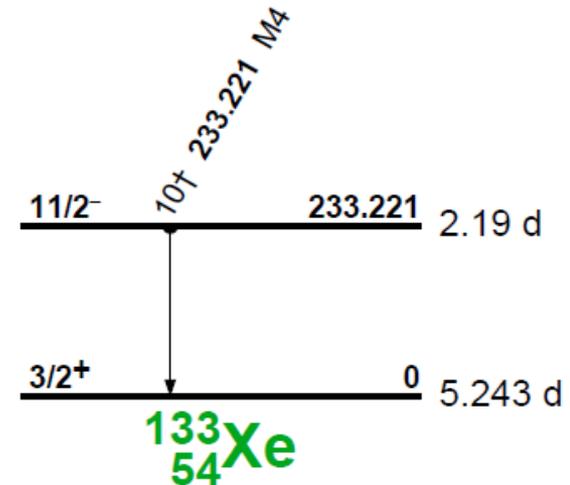
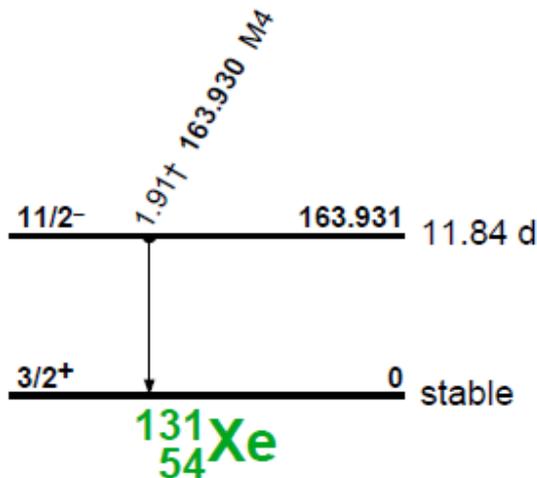
Y. Zheng, G.W. Miller, W.A. Tobias, G.D. Cates, Nature 537, 652 (2016)

- **$^{131\text{m}}\text{Xe}$** : $t_{1/2} = 12$ days
- **Setup**: low B-field
- **Results**: space-resolved signal (recorded pixel after pixel) with $1e^{13}$ nuclei vs $1e^{24}$ normally



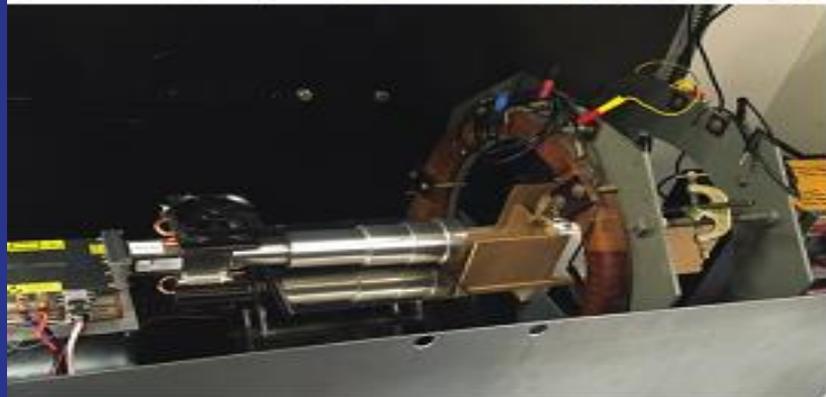
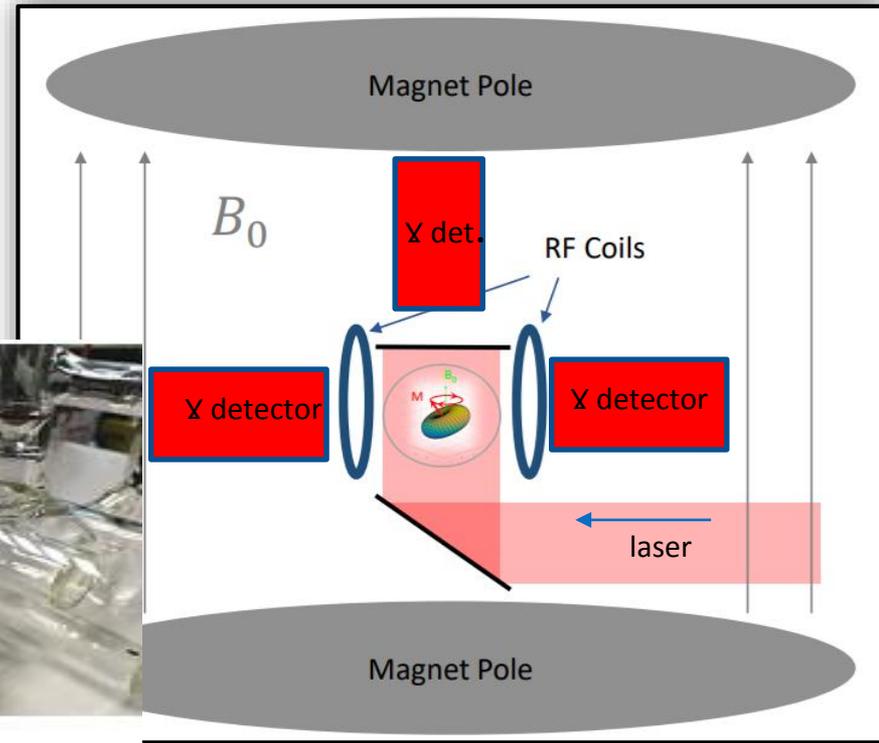
Our gamma-MRI project

- 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:
 - ^{133}mXe well produced at ISOLDE
 - Compact setup to polarize and detect asymmetry tested
- Next step: polarization of ^{133}mXe

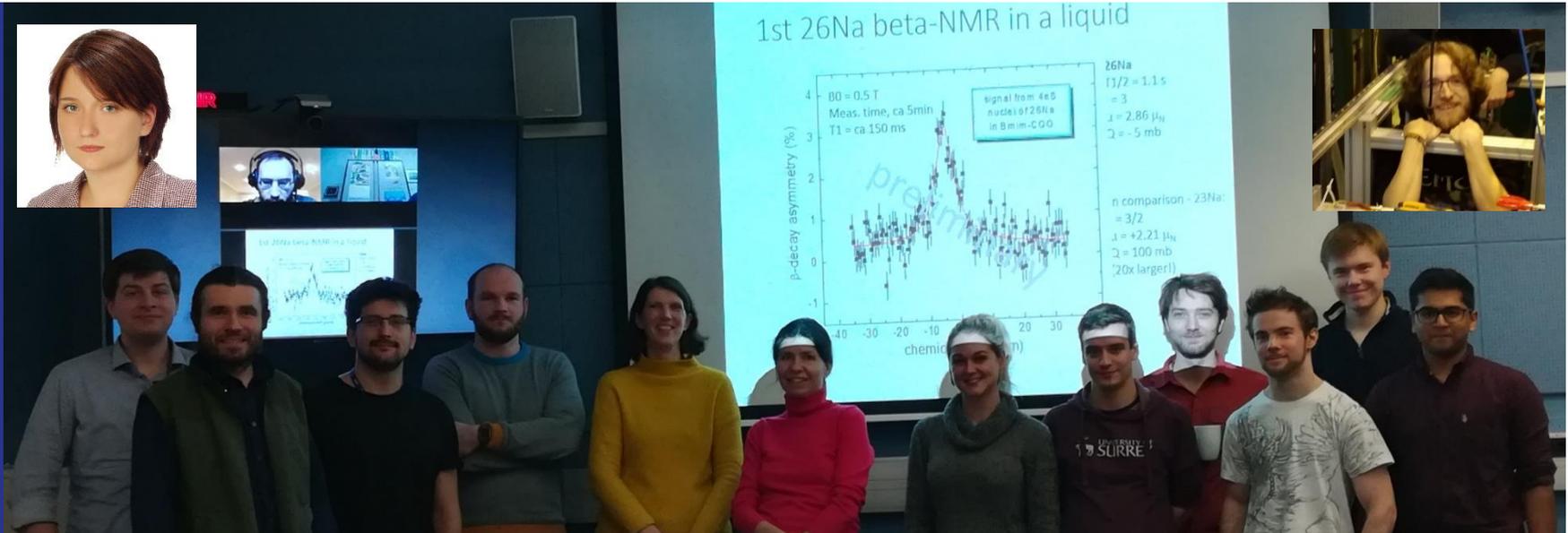


Summary and outlook

- Spin polarization seen in spatial distribution of nuclear decays:
- Beta decay asymmetry => V_{ud} 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

Acknowledgements

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Gamma-MRI:

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UNIVERSITÉ
DE GENÈVE



KU LEUVEN

IOOLDE

Vud

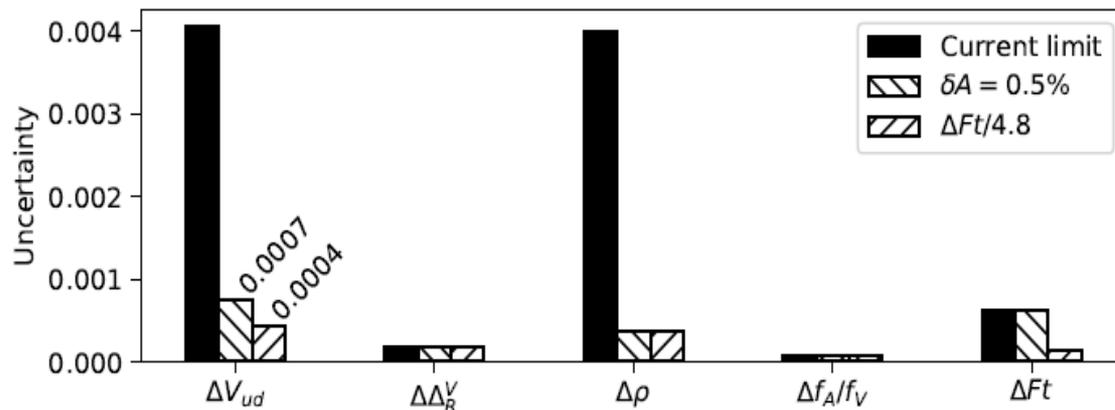
Assuming no Beyond Standard Model physics, the asymmetry parameter A_{SM} is given by [26]

$$A_{SM}^{\beta\mp} = \frac{\mp\lambda_{I_f, I_i}\rho^2 - 2\delta_{I_f, I_i}\sqrt{\frac{I_i}{I_i+1}}\rho}{1 + \rho^2}, \quad (1.7)$$

with

$$\lambda_{I_f, I_i} = \begin{cases} 1 & \text{for } I_f - I_i = -1 \\ \frac{1}{I_i+1} & \text{for } I_f - I_i = 0 \\ -\frac{I_i}{I_i+1} & \text{for } I_f - I_i = 1 \end{cases}. \quad (1.8)$$

35Ar:



Parameter	Situation 1	Situation 2
I_{dcy}	1×10^6	0.5×10^6
P	30%	20%
t	16 hours	3 days

Probe nuclei

Already polarized at ISOLDE

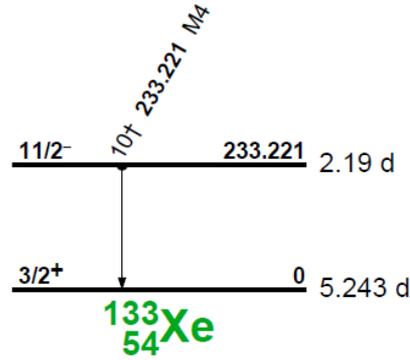
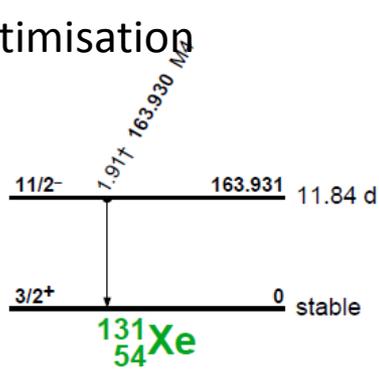
Nucleus	half-life	spin	magn mom (μ_N)	quadr mom (mb)	beta asym
8Li	0.84 s	2	1.65	31	5%
9Li	0.18 s	3/2	3.44	-31	
11Be	13.8	1/2	-1.68	0	1%
26Na	1.1 s	3	2.86	-5	30%
27Na	0.3 s	5/2	3.89	-7	30%
28Na	30 ms	1	2.43	40	40%
29Mg	1.2 s	3/2	0.98	160	3%
31Mg	0.25 s	1/2	-0.88	0	8%

Feasible and planned soon

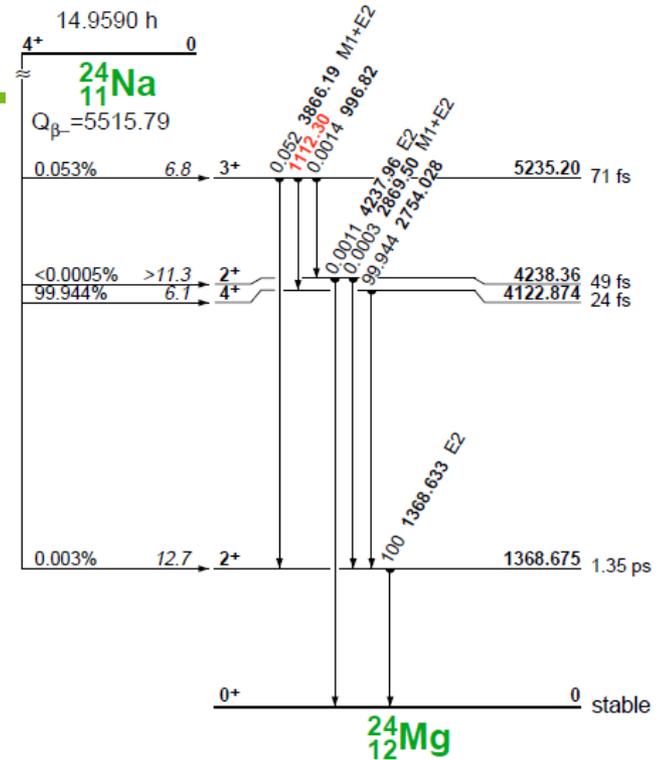
Nucleus	half-life	Nuclear spin	magn mom (μ_N)	Quadr mom (mb)
37K	1.2 s	3/2	+0.20	
49K	1.3 s	1/2	+1.34	0
39Ca	0.8 s	3/2	1.02	+38
51Ca	0.36 s	3/2	-1.05	+36
58Cu	3.2 s	1	0.57	-150
74Cu	1.6 s	2	-1.07	260
75Cu	1.2 s	5/2	1.01	-270
75Zn	10 s	7/2		
75mZn	5 s	1/2		0
77Zn	2 s	7/2		
77mZn	1.1 s	1/2		0

Our 1st isotopes

optimisation

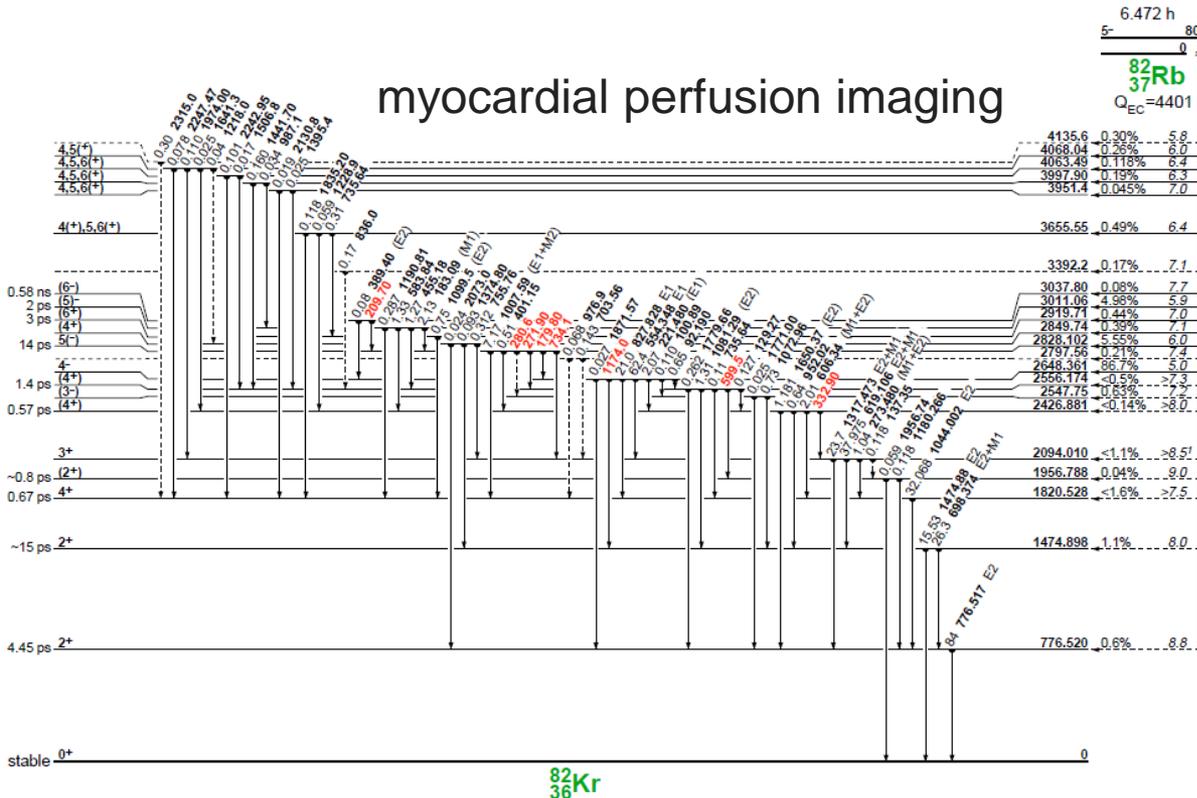


Ventilations studies



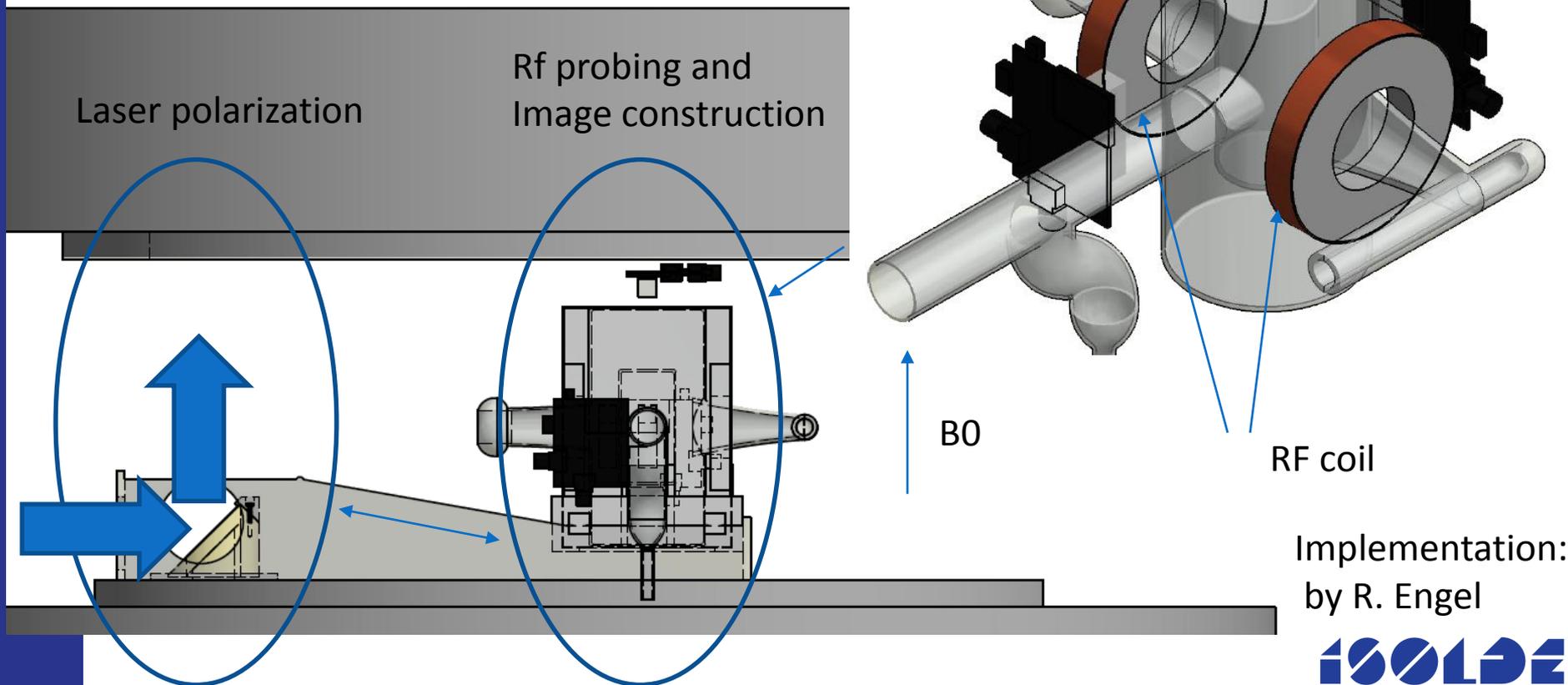
Studies of electrolytes

myocardial perfusion imaging



Our test gamma-MRI setup

- 0.1 T MRI magnet
- Glass vial with Rb and Xe
- Laser polarization inside strong field
- NaI detector crystals and Si PMTs



Vud

- comparative half-lives (ft values) are the same for different superallowed transitions:

- Ft depends on decay energy and partial half-life of a transition

- Leads to derivation of vector coupling constant

- Pion decay: $|V_{ud}| = 0.9728 \pm 0.0030,$

- Vus $|V_{us}| = 0.2248 \pm 0.0006.$

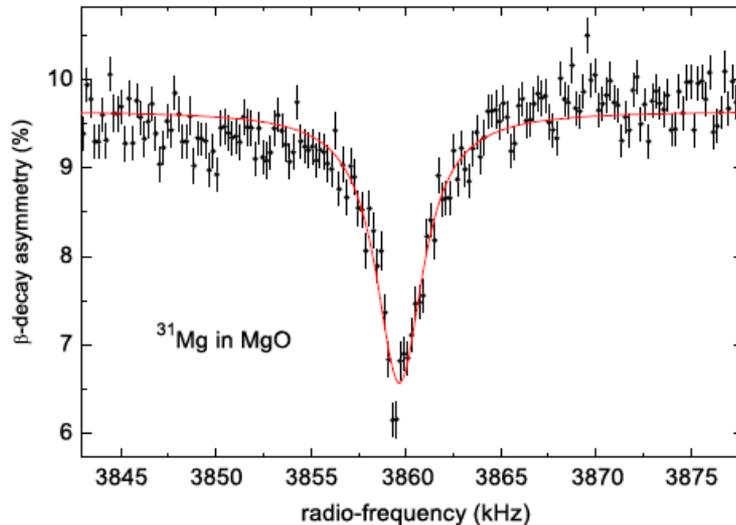
$$K_L^0 \rightarrow \pi e \nu, K_L^0 \rightarrow \pi \mu \nu, K^\pm \rightarrow \pi^0 e^\pm \nu, K^\pm \rightarrow \pi^0 \mu^\pm \nu \text{ and } K_S^0 \rightarrow \pi e \nu$$

- Vu $B \rightarrow X_u \ell \bar{\nu}$

$$|V_{ub}| = (4.49 \pm 0.16 \begin{matrix} +0.16 \\ -0.18 \end{matrix}) \times 10^{-3}.$$

Determining spins & moments

- Destroying polarization with RF (NMR):
 - When RF matches energy difference between spin up and down -> asymmetry disappears
 - ν_{RF} depends on spin, magnetic and quadrupole moment



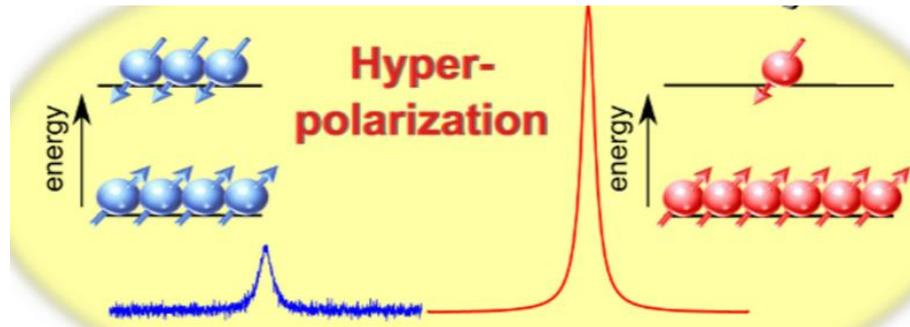
$$|g_I(^{31}\text{Mg})| = 1.7671(3)$$
$$I = \frac{1}{2}$$

⇒ 2 neutrons need to be promoted across N = 20:

-³¹Mg inside island of inversion
-understanding of inversion mechanism

Hyperpolarization

- Aligning almost all spins in one direction
 - => much higher degree of polarization than in thermal equilibrium
 - T1 relaxation time important: return to equilibrium
- => **Gain up to 10^5 in sensitivity**



- Hyperpolarization with laser light = optical pumping
- Works best for alkali and alkali earth metals
 - Strong and closed optical transitions in VIS or IR region

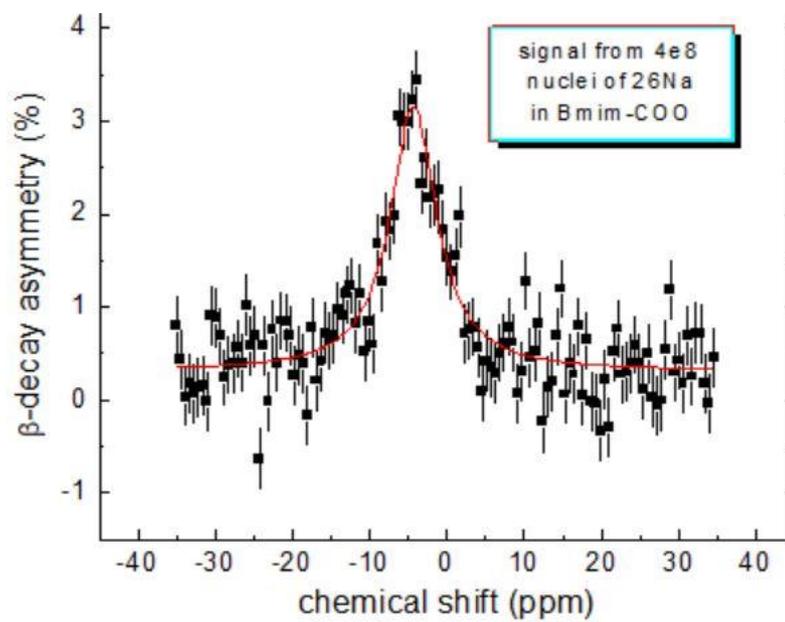
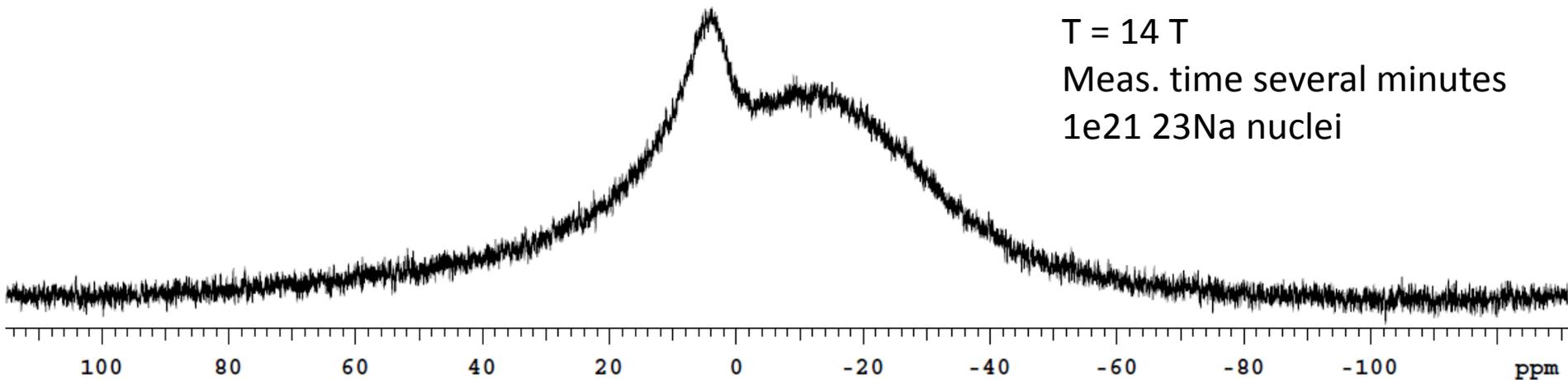
Comparison to conventional NMR

- ^{23}Na spectrum in Bmim-Ac ionic liquid (Bmim-COOH study ongoing)

T = 14 T

Meas. time several minutes

$1e^{21}$ ^{23}Na nuclei

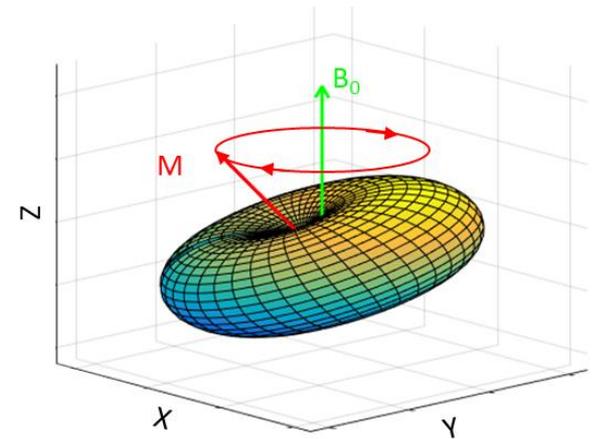


Gamma MRI – signal recording

- Magnetisation manipulated with RF pulses as in regular MRI
- Magnetization precesses around B_0 with Larmor frequency after irradiation with resonant RF pulse (e.g. $\pi/2$)
- precession registered as undulation of detector count-rates

=> this replaces detection of RF-signals in regular MRI

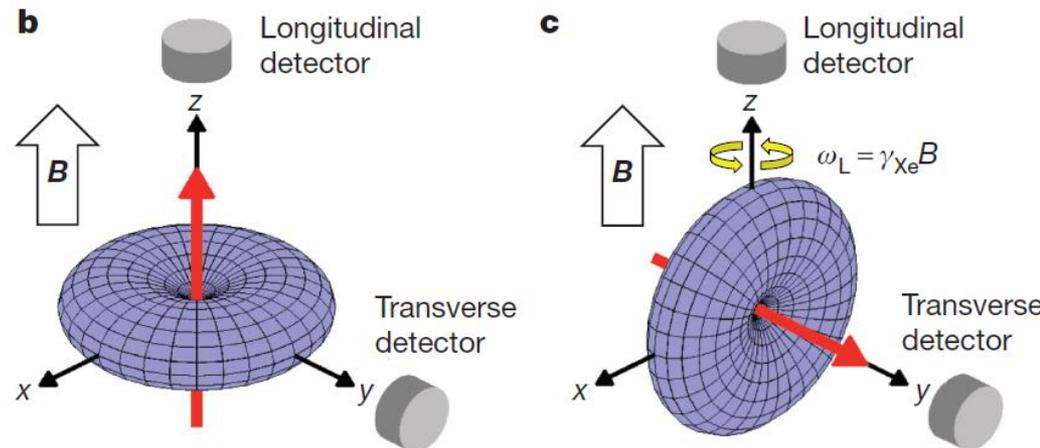
- When 1 pixel in resonance: change in gamma counts from all pixels visible by one detector

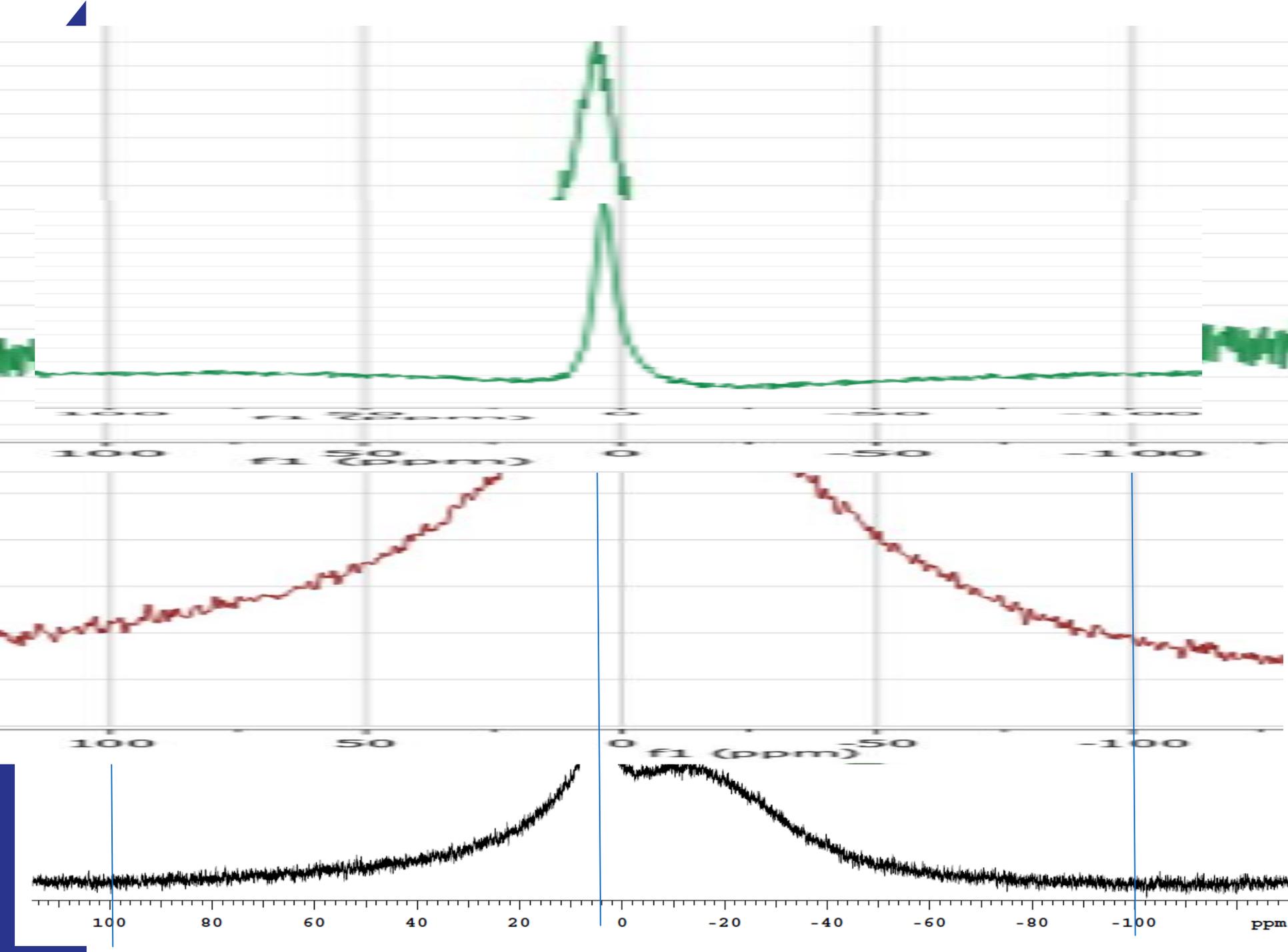


R. Engel

- **Strength:**
 - Up to $1e10$ higher sensitivity
 - -> trace amounts enough

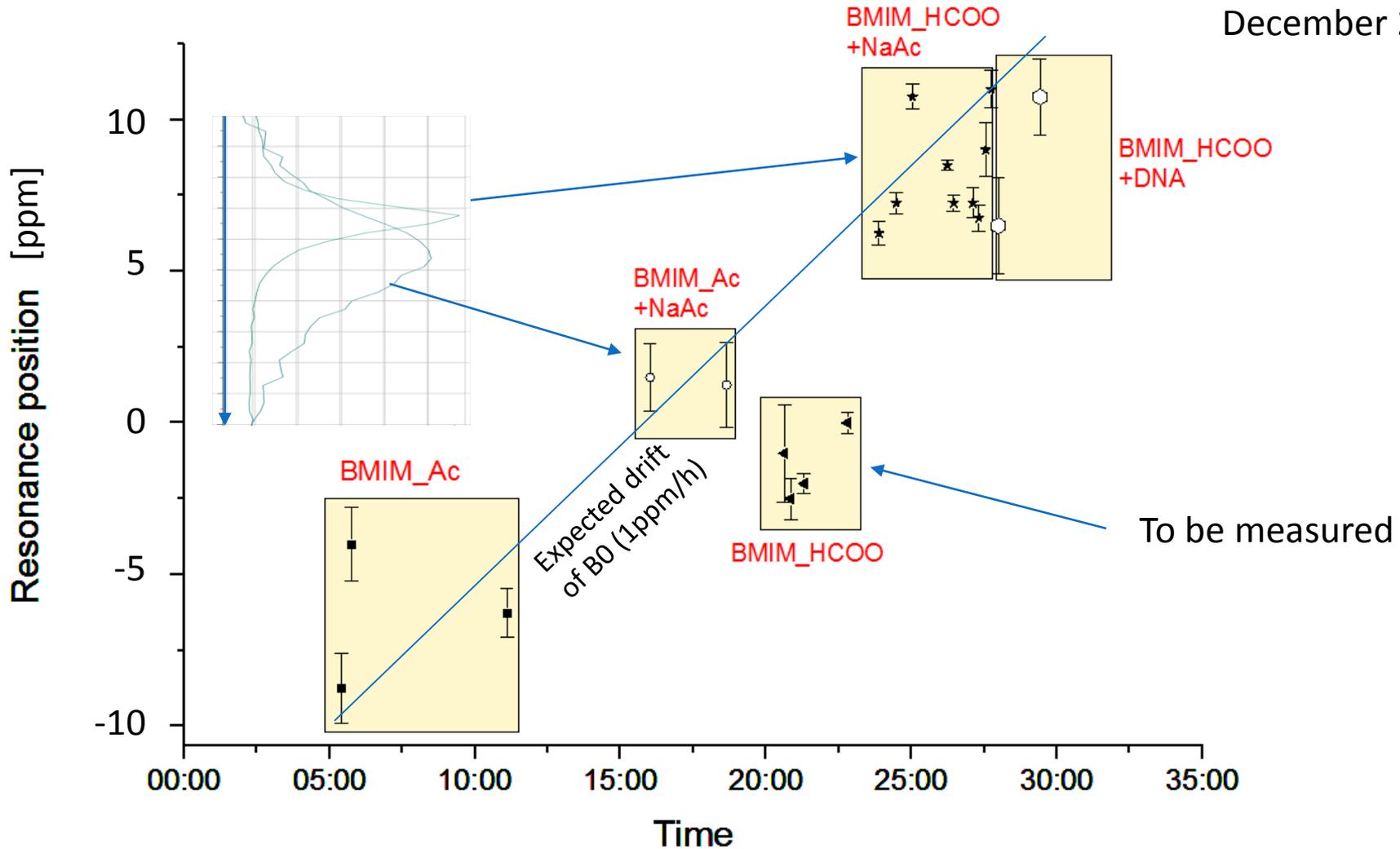
- **Drawback:**
 - Gamma signal from many pixels
 - Radioactive tracer





Latest results

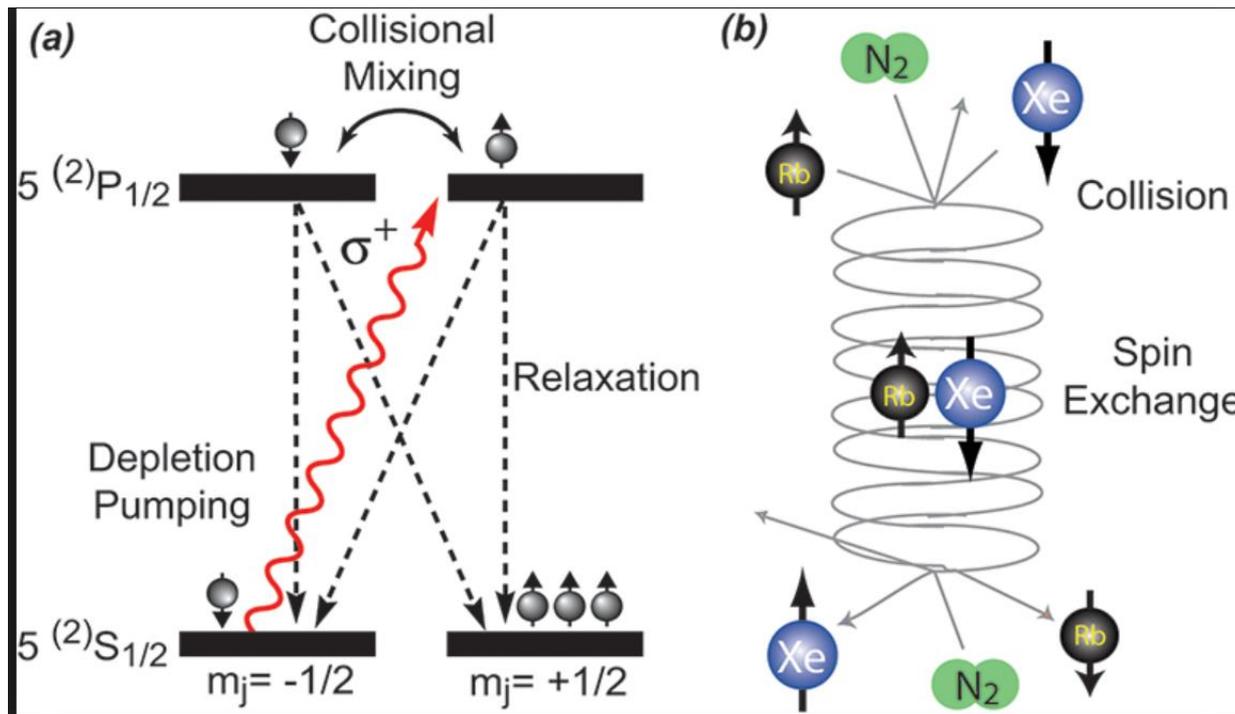
December 2017



In addition: conventional ^{23}Na studies performed 1.5 weeks ago

Spin-exchange optical pumping

- Multiple excitation cycles with circularly-polarized light
- Photon angular momentum transferred to valence electron
- Collisions with closed-shell atom transfer polarization to its nucleus
 - Works best for alkali atoms colliding with noble gas atoms
 - Nuclear spin-polarization of 10-90%
 - Noble gas polarization time transfer - minutes



Imaging

- The count rate after each CMZ pulse sequence represents the real or complex component of one point in the k-space.
- ◆ k-space location is encoded in the delay time τ and the Gradient strength G :

- ◆ The image is constructed from the assembled k-space data.

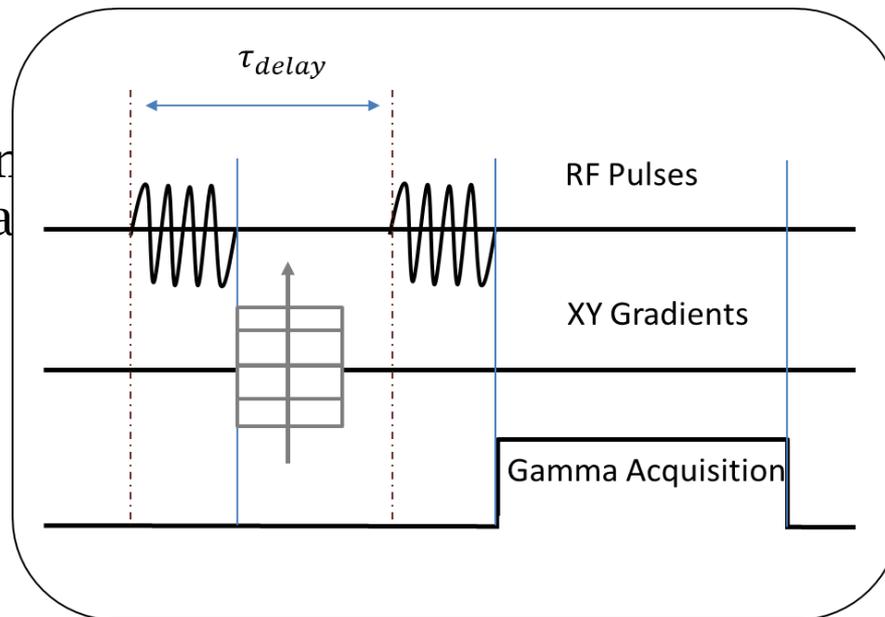


Figure 6: Imaging Pulse sequence after Cates, Miller & Zheng [2]

gammaMRI setup

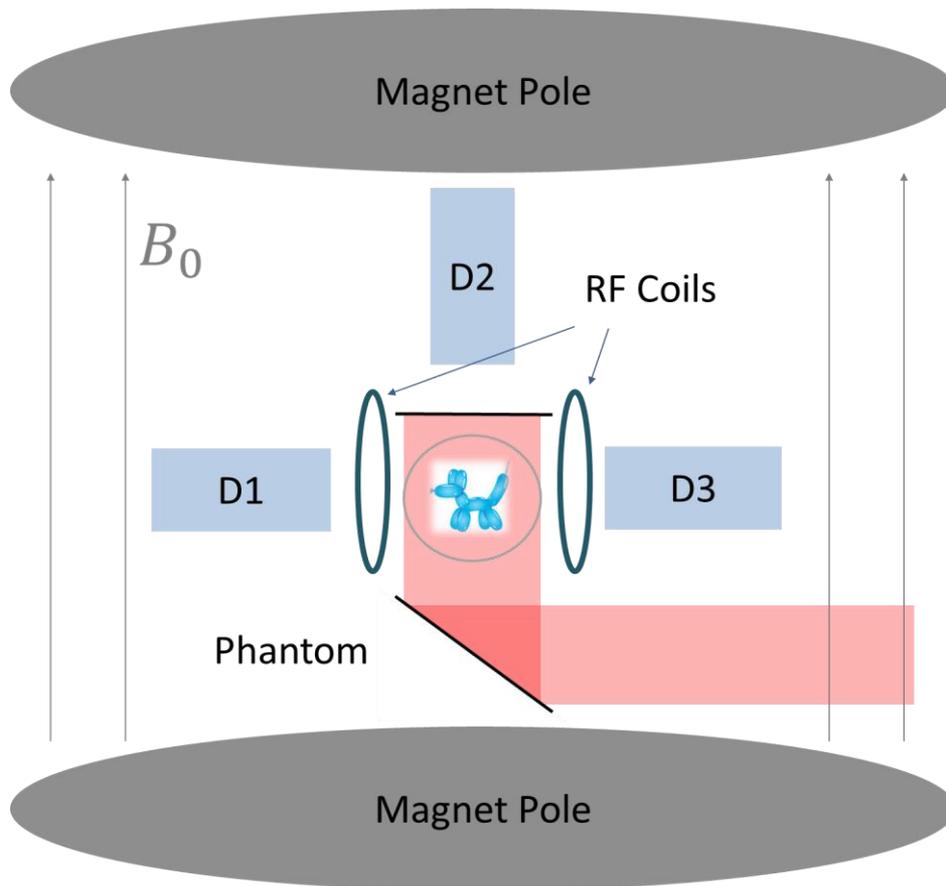
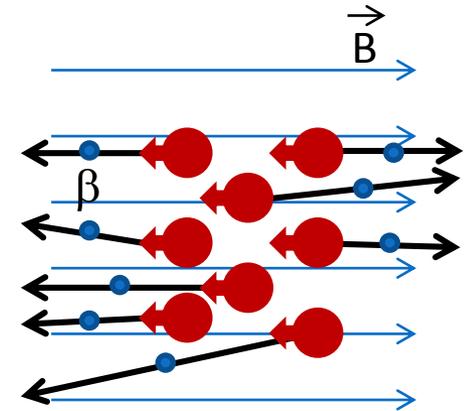


Figure 1: Schematic drawing of the planned setup. Gradient coils omitted for simplicity. D1-D3 represent gamma detectors.

Asymmetry in beta-particle emission

Weak interaction doesn't conserve parity:

- Anisotropic emission of beta particles from decay of polarized nuclei



Angular distribution of beta-radiation:

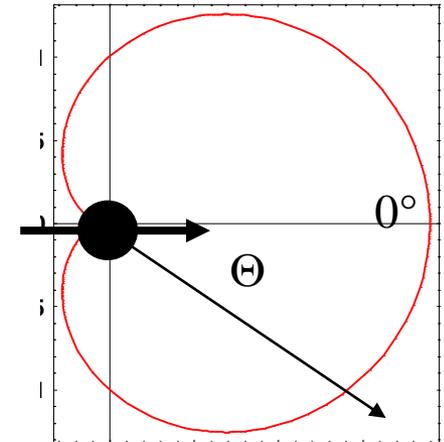
Velocity of beta-particle (v/c close to 1)

$$D(\theta) = 1 + a \frac{v}{c} \frac{\langle I_z \rangle}{I} \cos(\theta)$$

Angle between beta-particle emission and direction of spin polarization

Asymmetry factor $(-1,1)$, depends on details of beta decay

PI (0-100%): degree of spin polarization



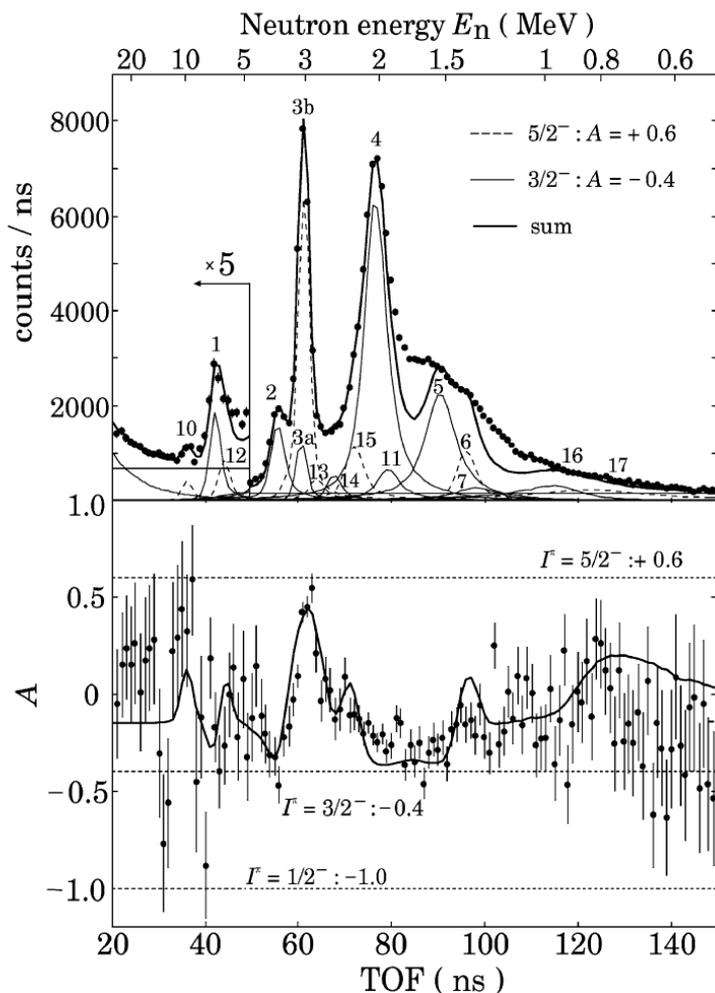
Measured β -decay asymmetry:
$$A = \frac{N(0^\circ) - N(180^\circ)}{N(0^\circ) + N(180^\circ)} = \frac{N_1 - N_2}{N_1 + N_2}$$

35Ar improvements

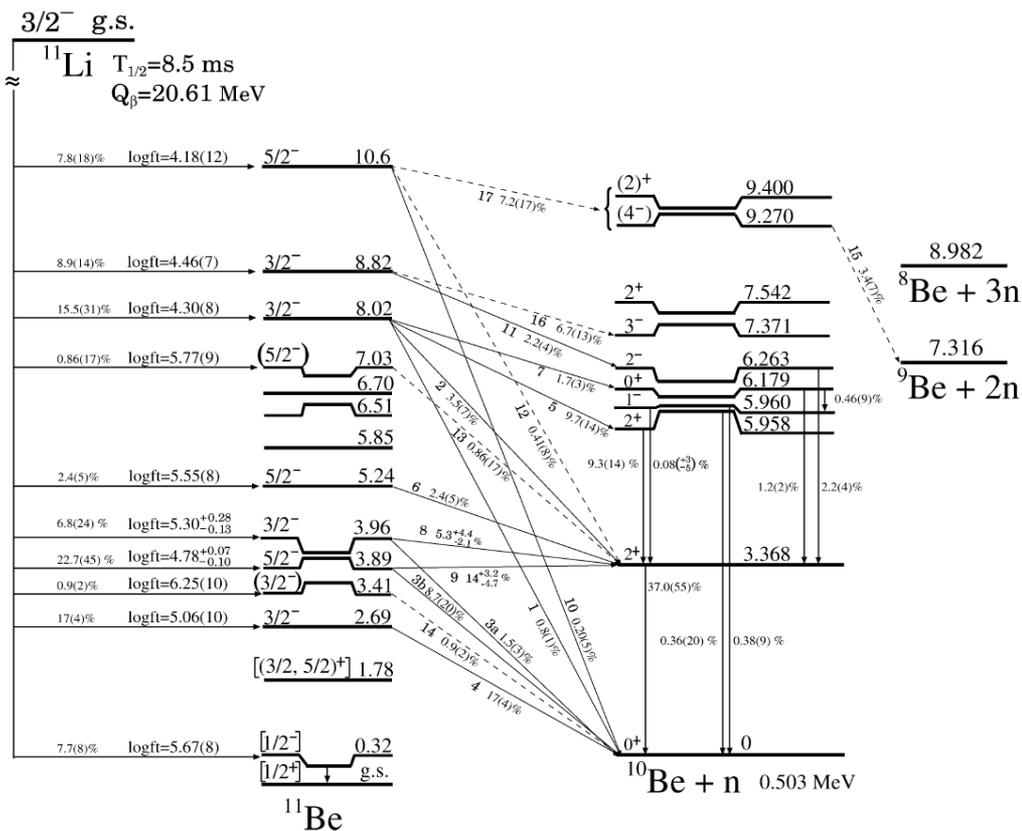
1. State-selective collisional re-ionization with Cl gas.
2. Selective re-ionization from the polarized metastable level using pulsed lasers.

Determining spins/parities of states

- a_β for a given transition depends on initial and final spins:
 - One of spins known + a_β measured => other spin is determined
 - Need to know degree of polarization (based e.g. on $a_\beta = 1$ transition)



=>

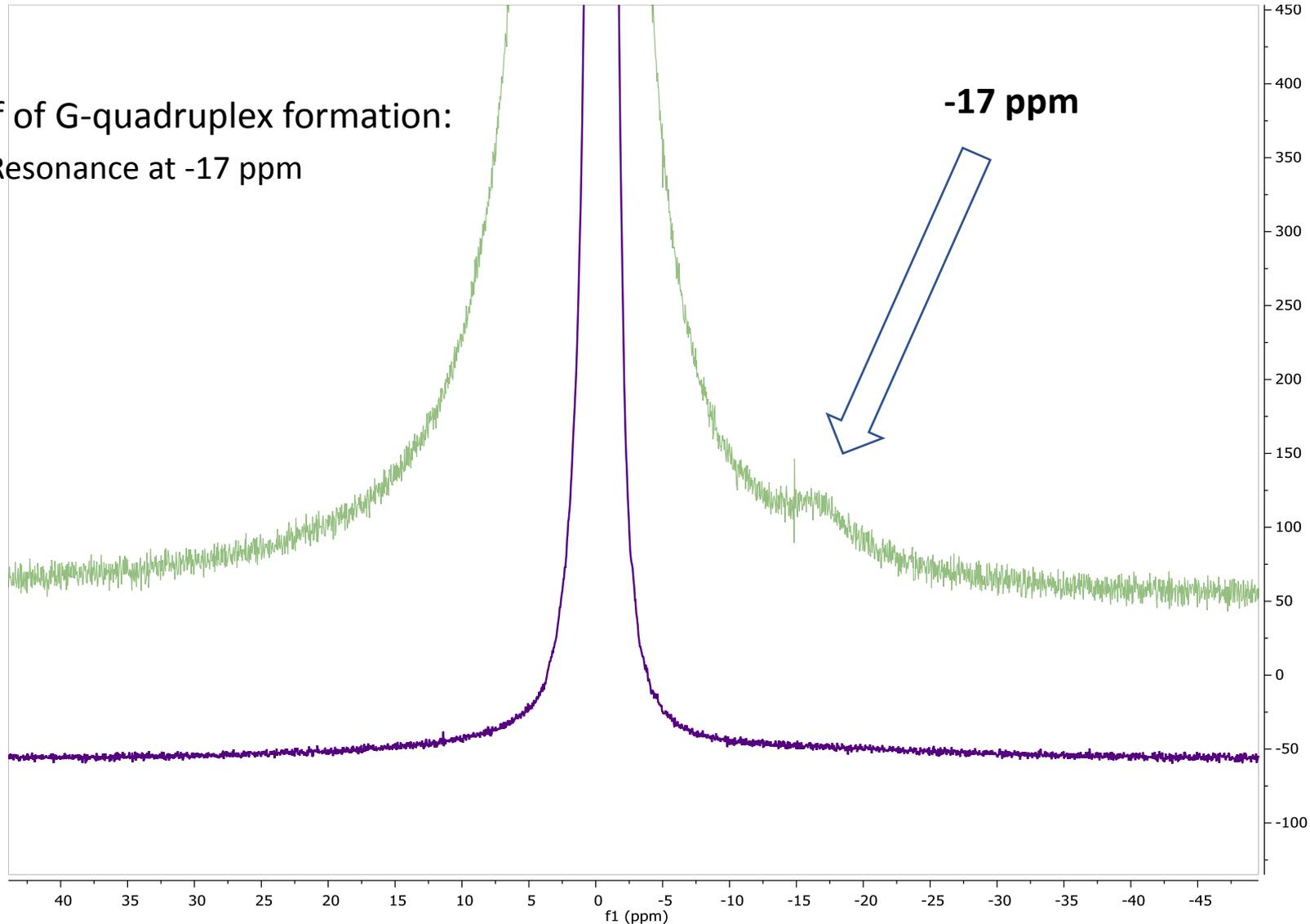


Laser polarization at TRIUMF

NMR on Na⁺ and G Quadruplexes

Conventional ²³Na NMR

- Proof of G-quadruplex formation:
 - Resonance at -17 ppm



0.7 M in H₂O

0.4 M in H₂O