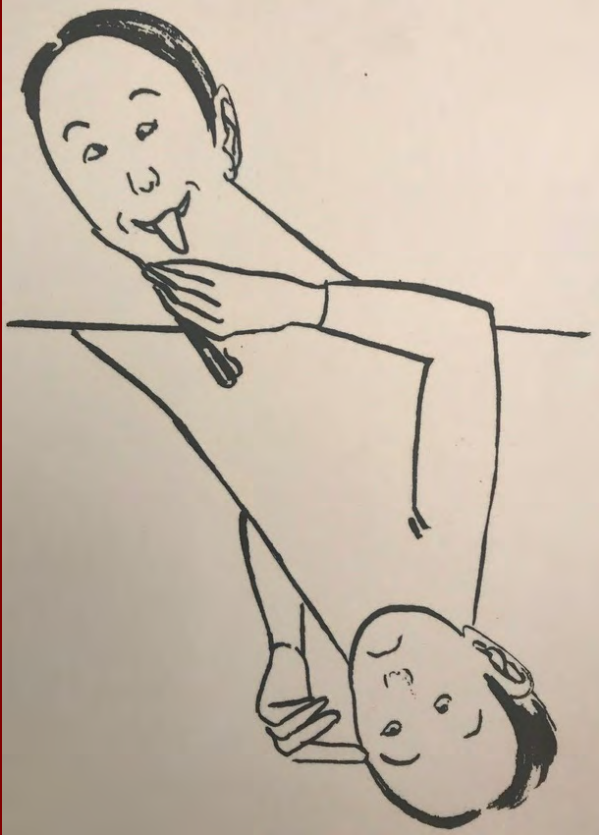


# Fundamental Physics with (Weird)

## Magnetic Resonance



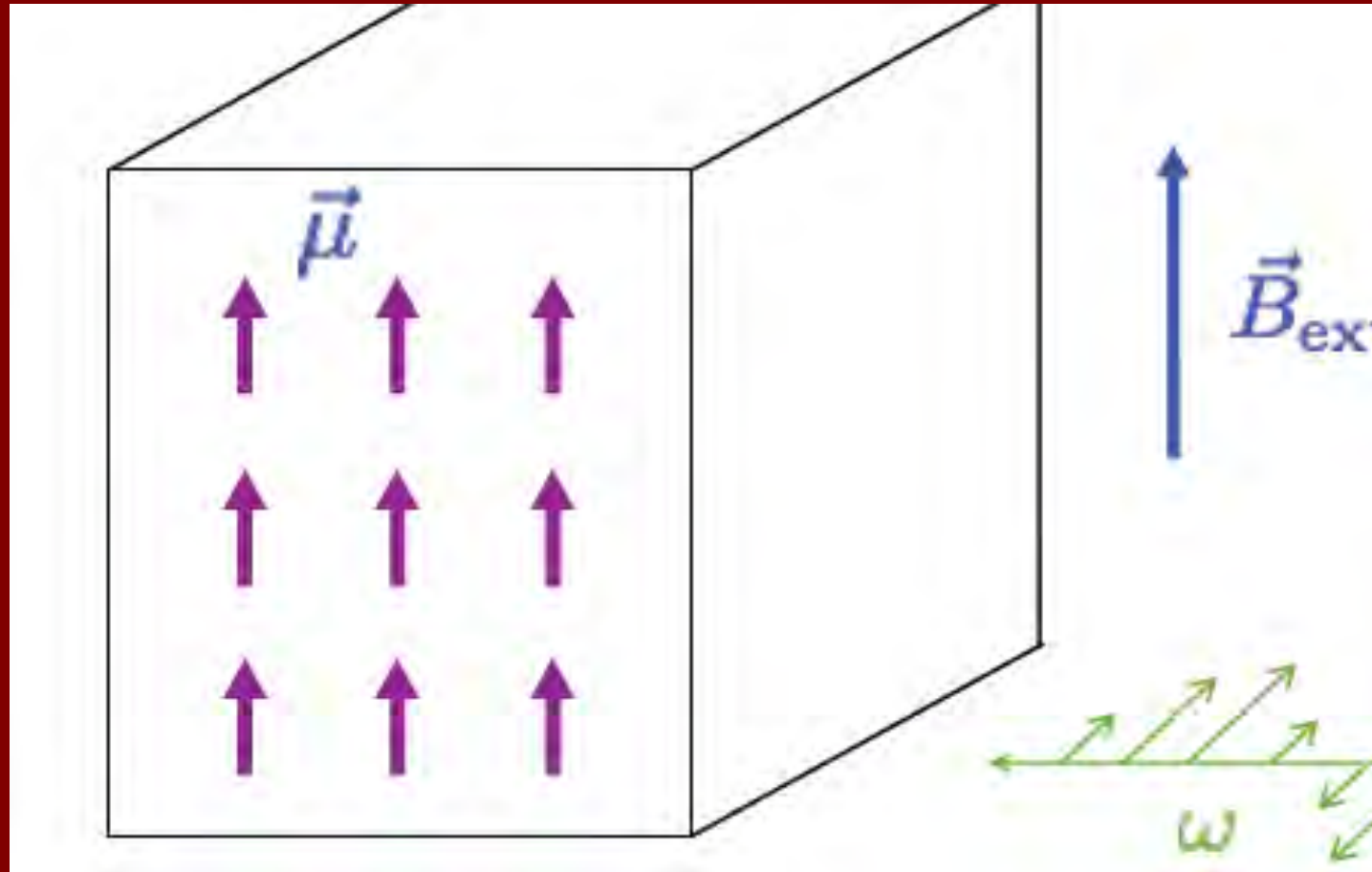
**Dmitry Budker**

**Helmholtz Institute  
JGU Mainz**

**UC Berkeley Physics  
NSD LBNL**

*Frascati, November 29, 2017*

# SEARCHING FOR ULTRALIGHT DARK MATTER WITH nuclear magnetic resonance



# So what is **DM** or what mimics it ?

- ▣ A gross misunderstanding of gravity (MOND, ...) ☹️?
- ▣ Proca MHD (finite photon mass) ?
- ▣ Black holes, dark planets, interstellar gas, ... ☹️
- ▣ WIMPS 😊
- ▣ Ultralight bosonic particles
  - **Axions** (pseudoscalar) 😊 ←
  - **ALPs** (pseudoscalar) 😊 ←
  - **Dilatons** (scalar) 😊
  - **Vector particles** 😊 ←
  - Tensor particles ???

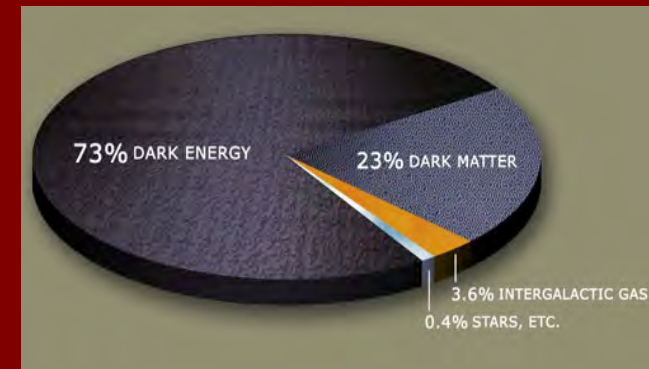
# “Most Wanted” file on DM

## What do we know?

- ▣ Galactic DM density:  $\sim 0.4 \text{ GeV/cm}^3$  (10 GeV/cm<sup>3</sup> d.g.)
- ▣ Has to be nonrelativistic:  $v/c \sim 10^{-3}$  (cold DM)
- ▣ Has to be **bosonic** if  $m < \sim 20 \text{ eV}$  (1 keV dwarf galaxies)
- ▣ “Bosonic Oscillator” with  $Q \sim (v/c)^{-2} \sim 10^6$
- ▣ Cannot be lighter than  $\sim 10^{-22} \text{ eV}$
- ▣ ... (e.g., BEC ?)

# Why Axions (ALPs) ?

- Big clean-up ?
  - Strong CP problem
  - Dark Matter
  - Dark Energy
  - Baryon asymmetry of the Universe
  - Hierarchy?
  - ...



<http://earthsky.org/space/>

# How to search for Axions (ALPs) ?

## Axion (ALP) Interactions

Gravity

P. Graham  
S. Rajendran

2017  
New Horizons  
In Physics Prize

Gauge Fields

Fermions

$$\frac{a}{f_a} F_{\mu\nu} \tilde{F}^{\mu\nu}$$

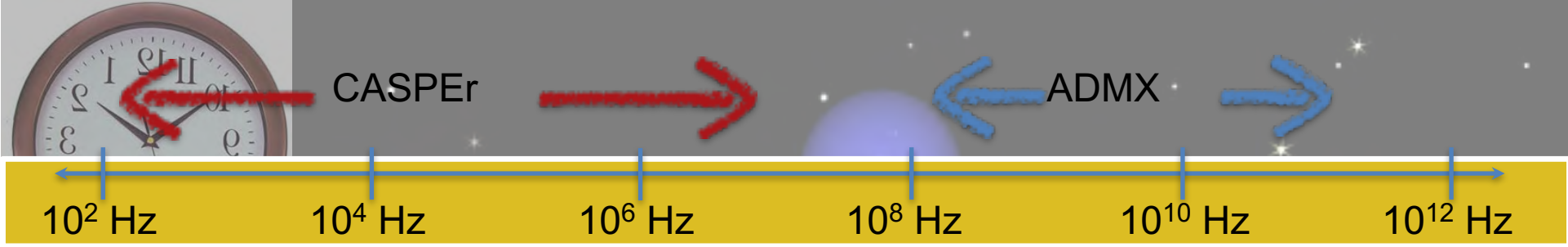
$$\frac{a}{f_a} G_{\mu\nu} \tilde{G}^{\mu\nu}$$

$$\frac{\partial_\mu a}{f_a} \bar{\Psi}_f \gamma^\mu \gamma_5 \Psi_f$$

Most  
Searches

(CASPER-**E**)

(CASPER-**Wind**, **GNOME**, QUAX)



axion mass (frequency)

# Cosmic Axion Spin Precession Experiment (CASPER)

Proposal:

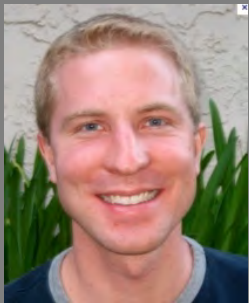
**Peter Graham**

**Surjeet Rajendran**

**Alex Sushkov**

**Micah Ledbetter**



**Dmitry Budker**



P. Graham & S. Rajendran PRD **88** (2013) arXiv:1306.6088,  
D. Budker *et al* PRX (2014) arXiv:1306.6089

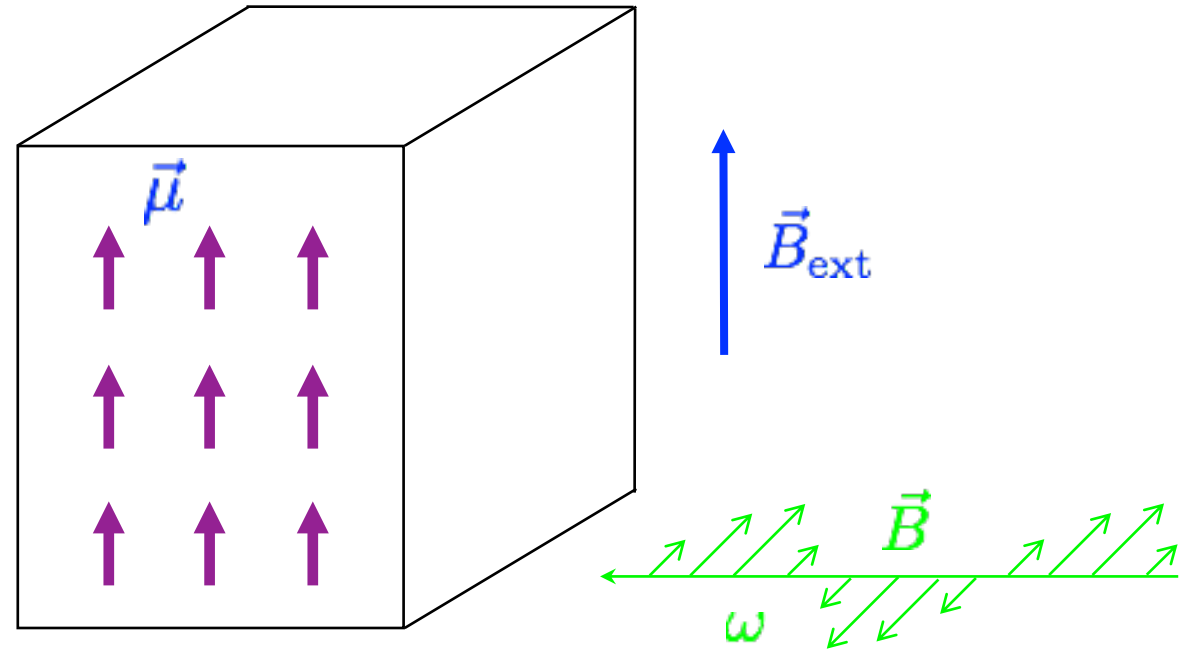
# CASPEr Overview

Key ideas:

- Axion (ALP) field **oscillates**
- at a frequency equal to its mass (mHz to GHz)
- → **time varying** CP-odd nuclear moments:
- nEDM, Schiff, ... 
- Also: **axion wind** (like a magnetic field)
- $v \sim 10^{-3} c$  (virial velocity) 
- Coherence time:  $[m_a(v/c)^2]^{-1} \rightarrow Q \sim 10^6$

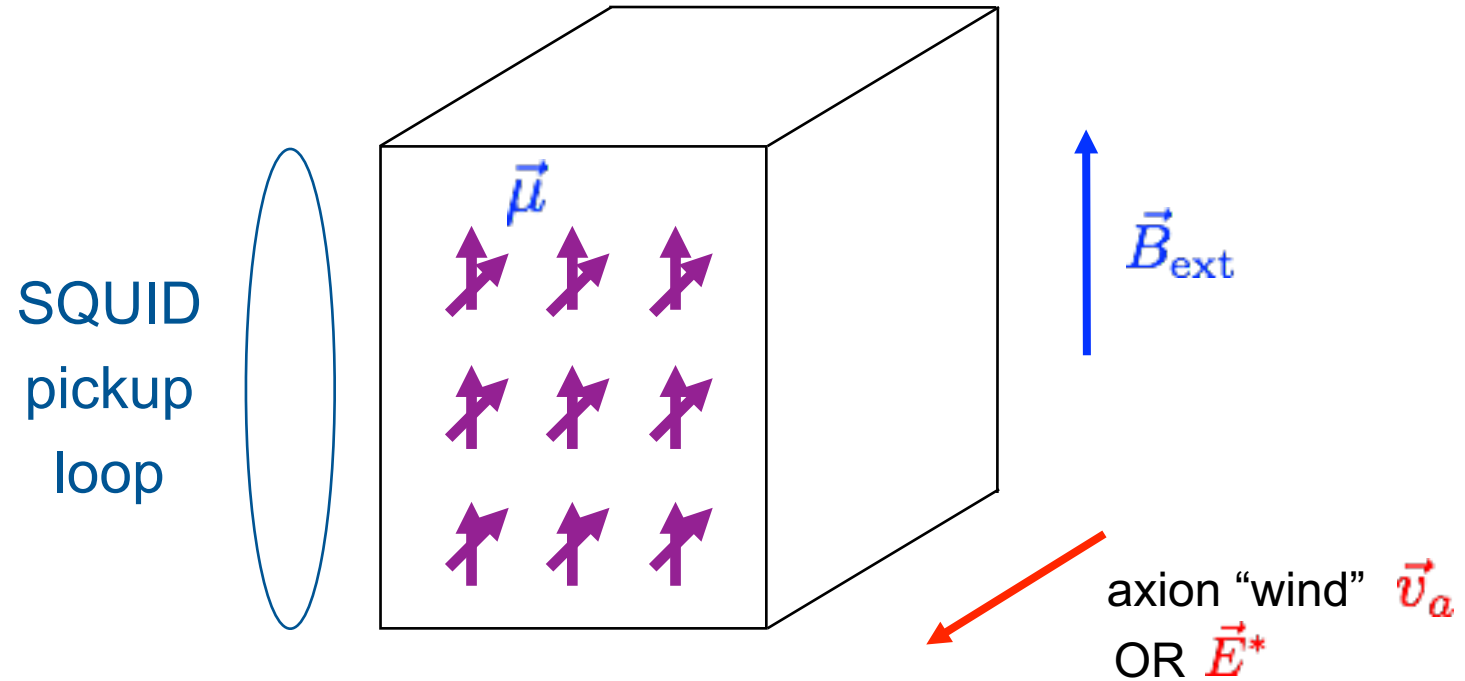


# Nuclear Magnetic Resonance (NMR)



Resonance:  $2\mu B_{\text{ext}} = \omega$

# CASPEr

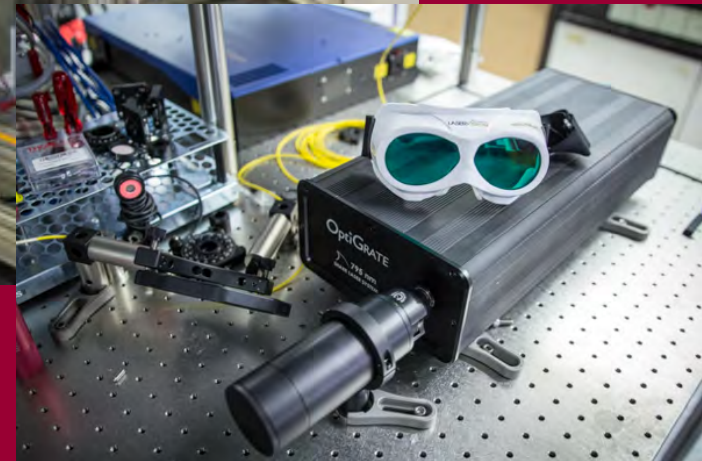


Larmor frequency = axion mass  $\rightarrow$  resonant enhancement

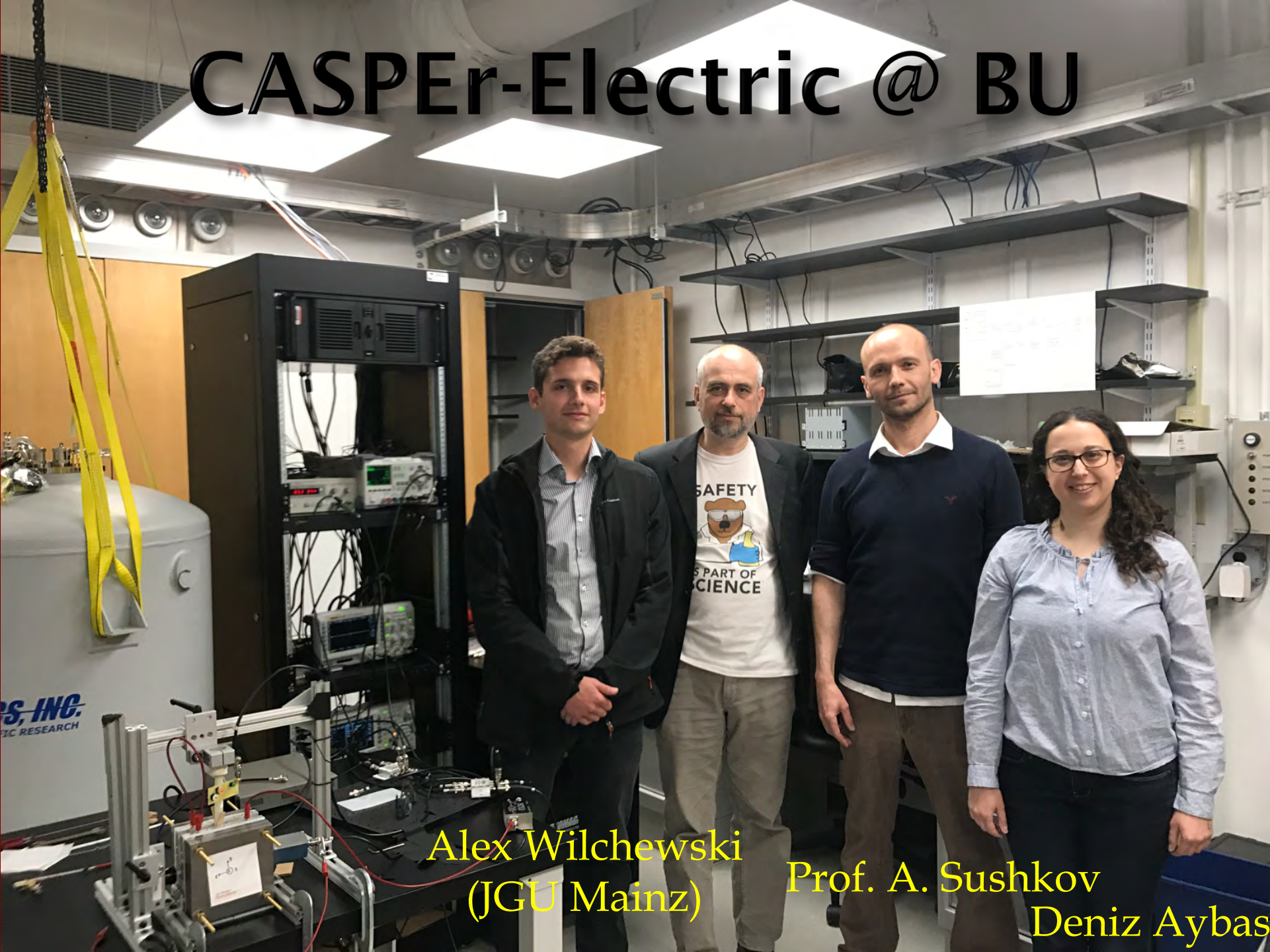
SQUID measures resulting transverse magnetization

Example materials: liquid  $^{129}\text{Xe}$ , ferroelectric  $\text{PbTiO}_3$

# Xe hyperpolarizer @ Mainz



# CASPER-Electric @ BU

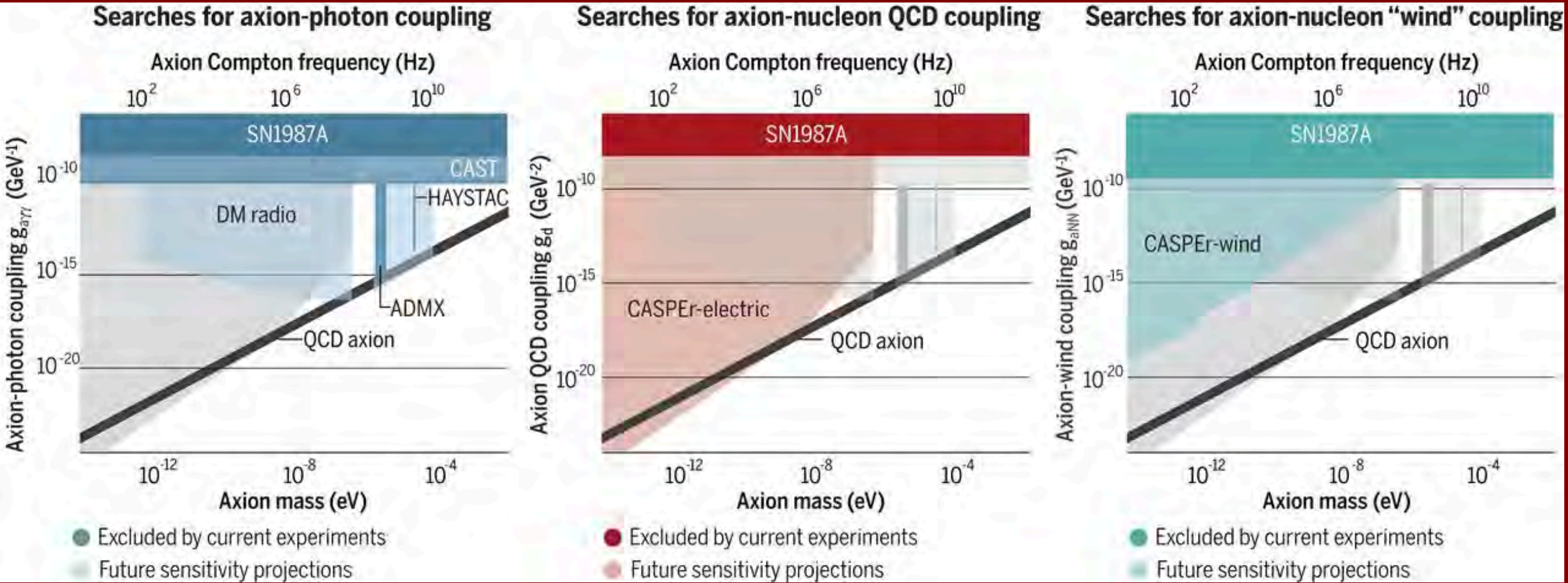


Alex Wilchewski  
(JGU Mainz)

Prof. A. Sushkov

Deniz Aybas

# Experimental constraints and projected sensitivities of axion dark-matter searches



# ZERO-FIELD

## nuclear magnetic resonance

Micah P. Ledbetter and Dmitry Budker

Counter to intuition, one doesn't necessarily need a strong magnet—or any magnet, for that matter—to extract richly informative spectra from nuclear spins.



April 2013 Physics Today

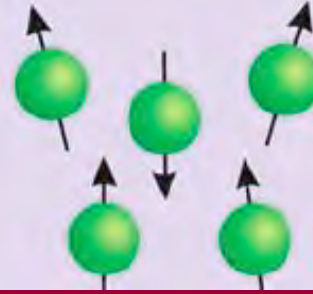
[www.physicstoday.org](http://www.physicstoday.org)

Micah Ledbetter

# Three Stages of NMR

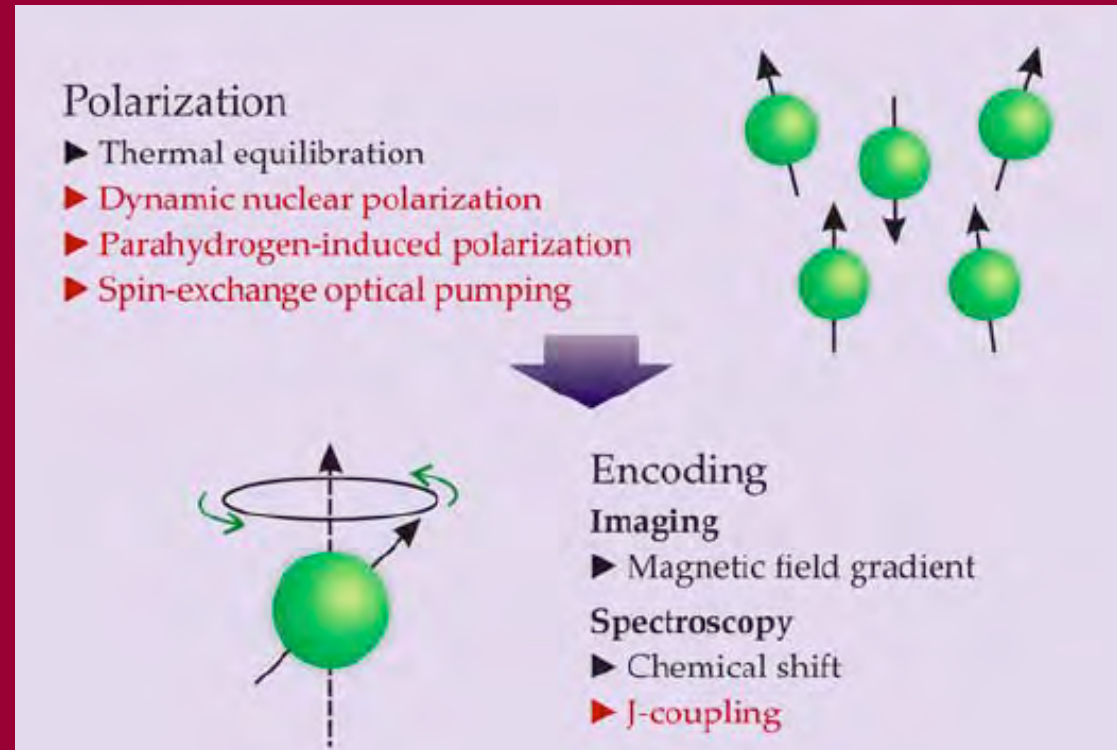
## Polarization

- ▶ Thermal equilibration
- ▶ Dynamic nuclear polarization
- ▶ Parahydrogen-induced polarization
- ▶ Spin-exchange optical pumping

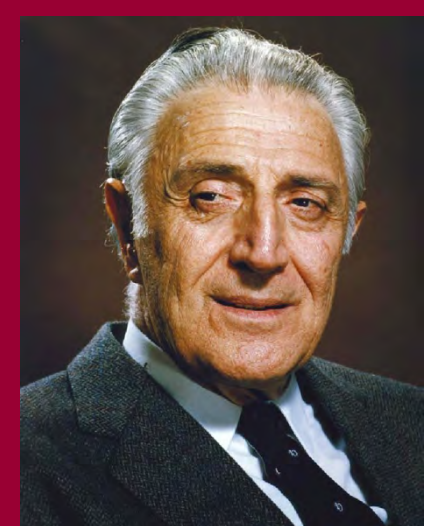


No need to polarize in  
spin-noise  
spectroscopy  
→ small N

# Three Stages of NMR



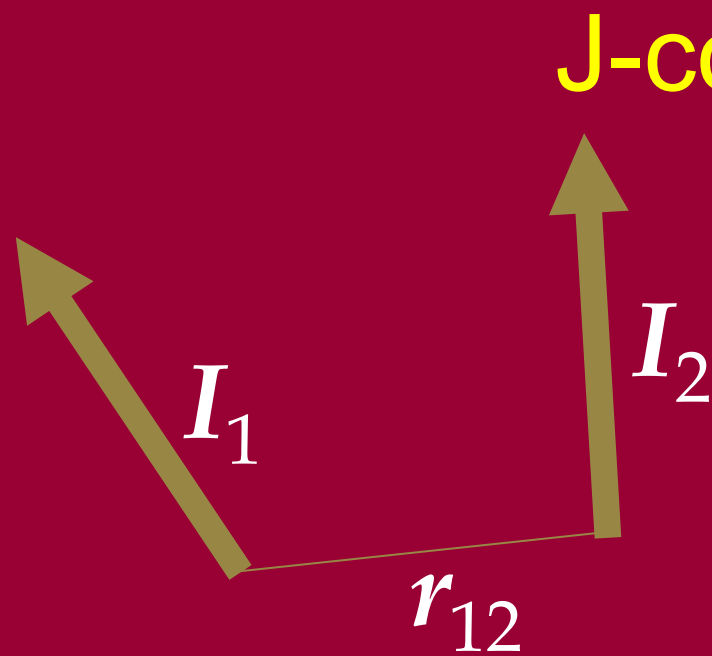




Erwin L. Hahn



C. P. Slichter



Dipole-dipole interaction

$$H \propto \frac{I_1 \cdot I_2}{r_{12}^3} (1 - 3\cos^2\theta)$$

averages by tumbling

J-coupling

$$H = JI_1 \cdot I_2$$

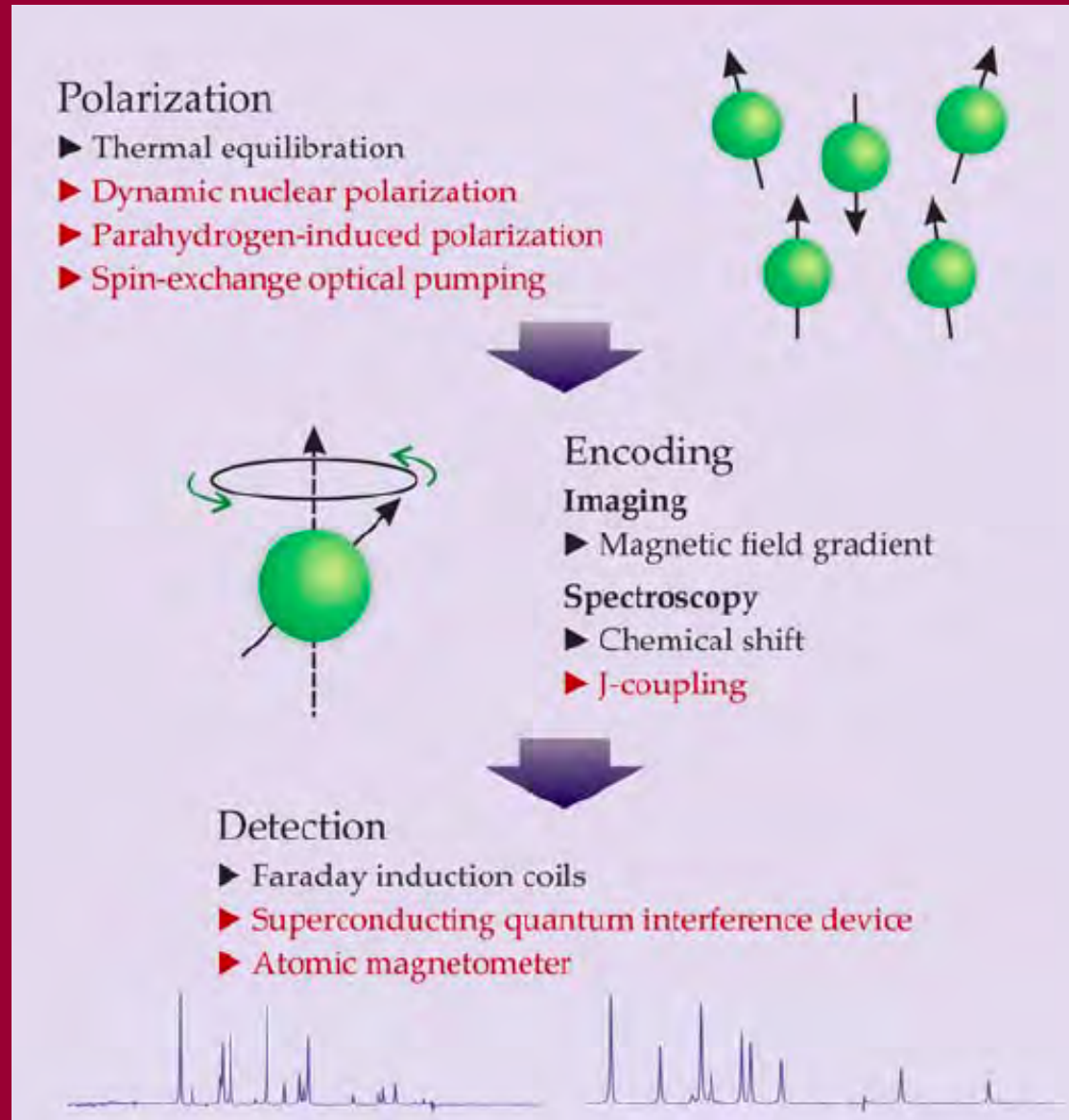
survives tumbling!

☞ second-order hyperfine

**Hahn, E.L.** & Maxwell, D.E. *Phys. Rev.* **84** 1246-1247 (1952)

Gutowsky, H.S., McCall, D.W., & **Slichter, C.P. J.** *Chem. Phys.* **21**, 279-292 (1953)

# Three Stages of NMR



# Parahydrogen induced polarization (PHIP)



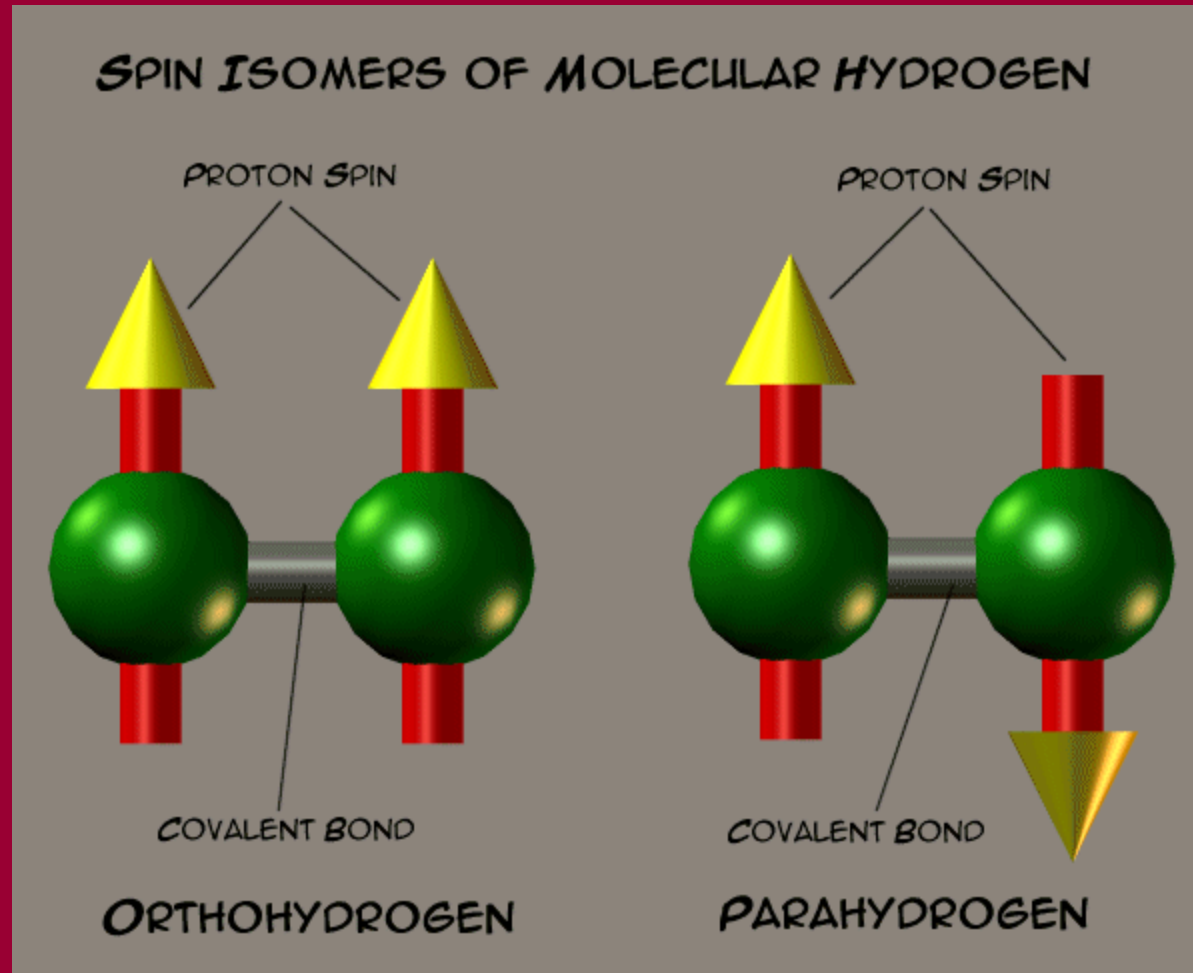
C. R. Bowers



Daniel P. Weitekamp

Transformation of symmetrization order to nuclear-spin magnetization  
by chemical reaction and nuclear magnetic resonance  
*PRL* **57** (21): 2645–2648 (1986)

# Parahydrogen 101

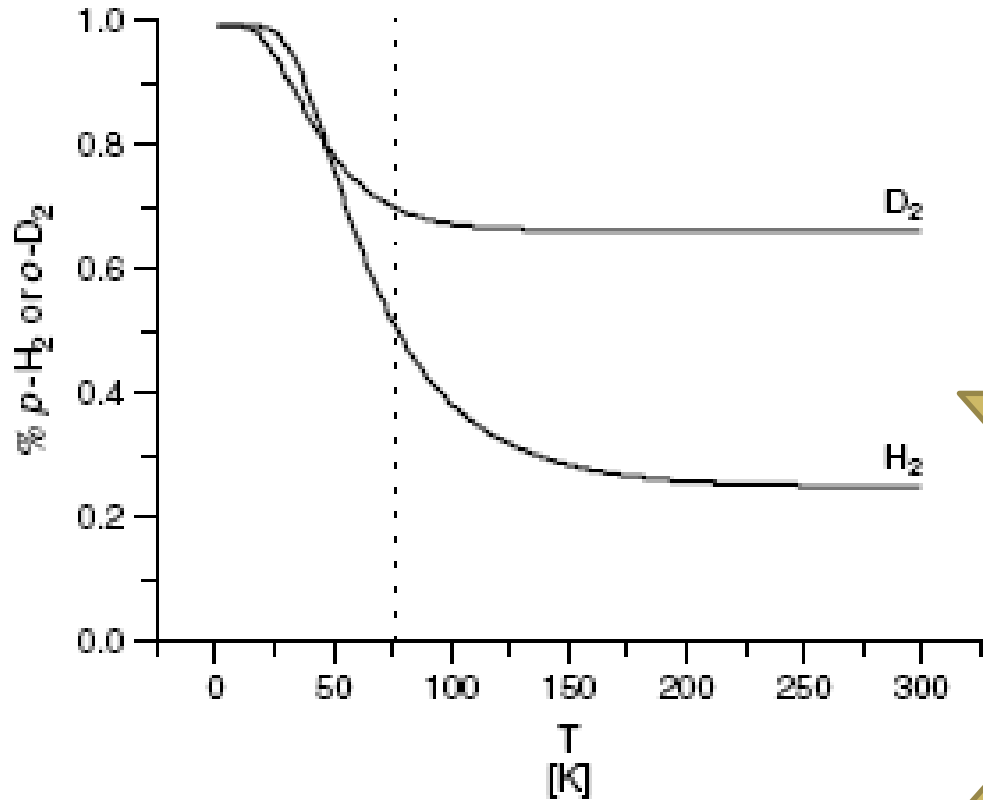


Odd J

Even J

# Parahydrogen 102

$$\frac{E_{J=1} - E_{J=0}}{k_B} = 2\theta_{rot} = \frac{\hbar^2}{k_B I} = 174.98 \text{ K}$$

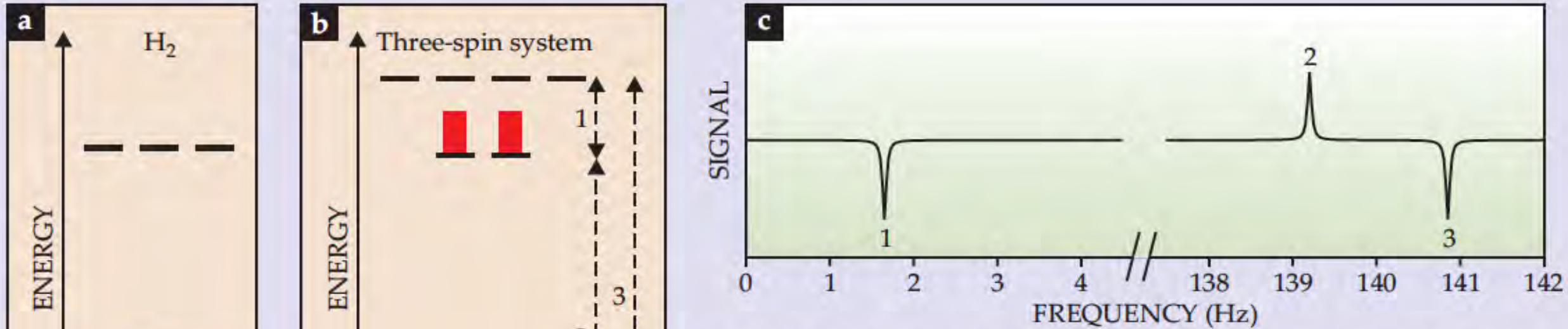


51% para @ 77K

99.9% para @ 4K

Spin-Statistics  
in action!

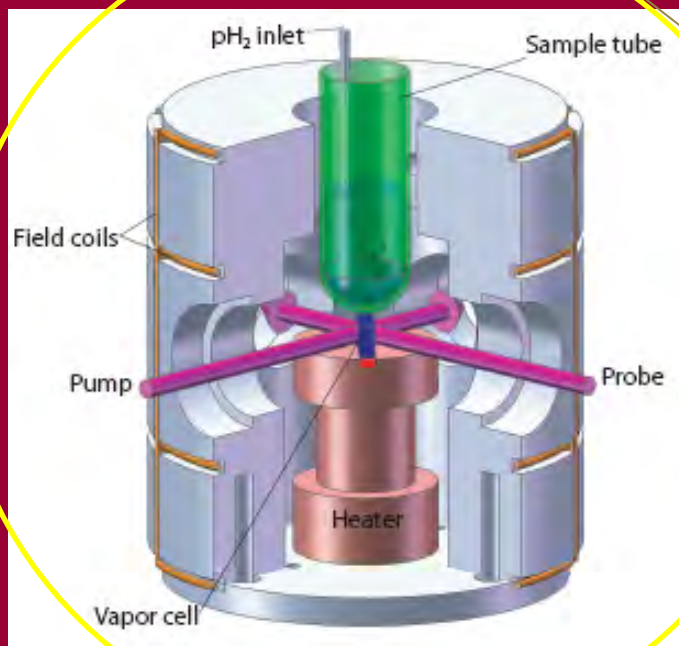
# Parahydrogen Induced Polarization



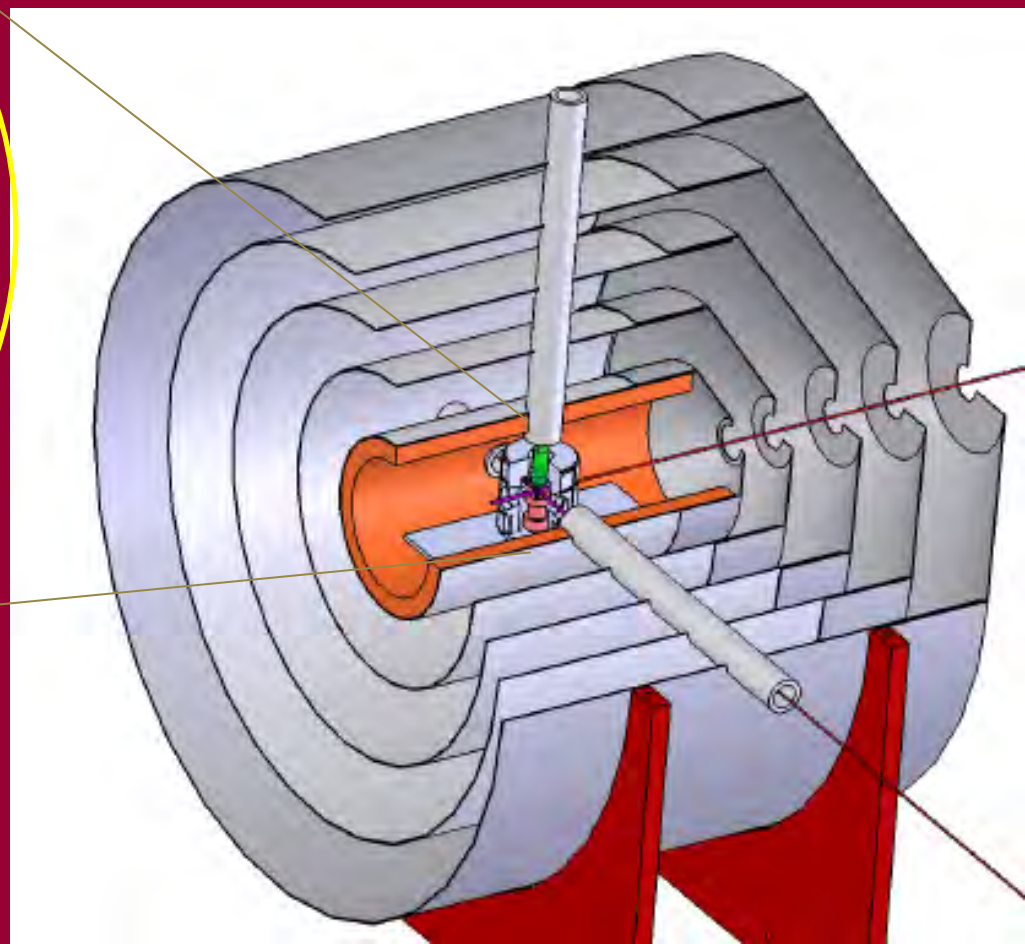
**Figure 5. Parahydrogen-induced polarization** can be used to obtain nuclear magnetic resonance signals in the absence of a magnetic field, as depicted here for a hypothetical three-spin system consisting of a carbon-13 nucleus and the nuclei of a parahydrogen molecule. **(a)** In isolation, the antiparallel spins in the parahydrogen molecule correspond to the singlet state. **(b)** If the molecule is catalytically added to a substrate

molecule containing  $^{13}C$ , and if one of the C–H couplings is much stronger than the other couplings in the system, the symmetry of the parahydrogen spins is broken and in the newly formed three-spin system, the population of the upper doublet is about three times that of the lower one. (Here, we ignore the rotational energies that may be correlated with the nuclear state.) The horizontal lines represent magnetic sublevels and the red rectangles represent the expected populations in each sublevel. **(c)** The simulated spectrum of a system with strong C–H coupling  $J_{CH} = 140$  Hz, weak C–H coupling  $J_{CH} = -5.2$  Hz, and H–H coupling  $J_{HH} = 7.7$  Hz yields the three peaks shown here, which correspond to the three allowed transitions indicated by the dashed arrows in panel b.

# NMR inside-out: $p\text{H}_2$ polarization; laser-mag detection

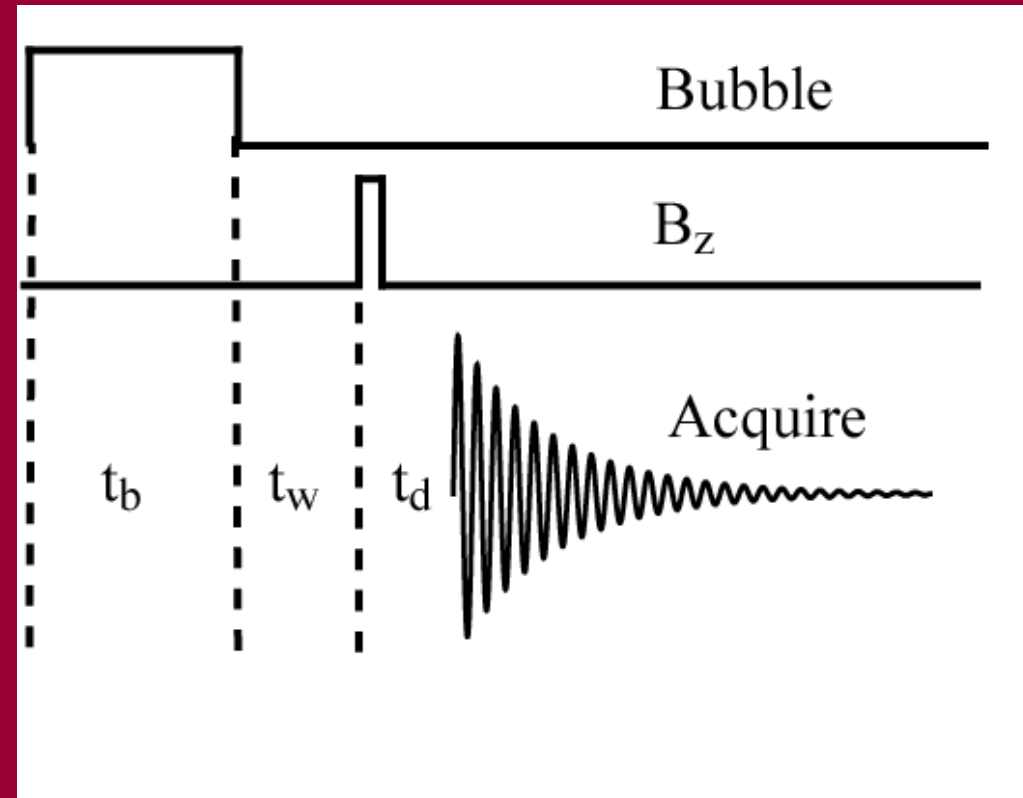
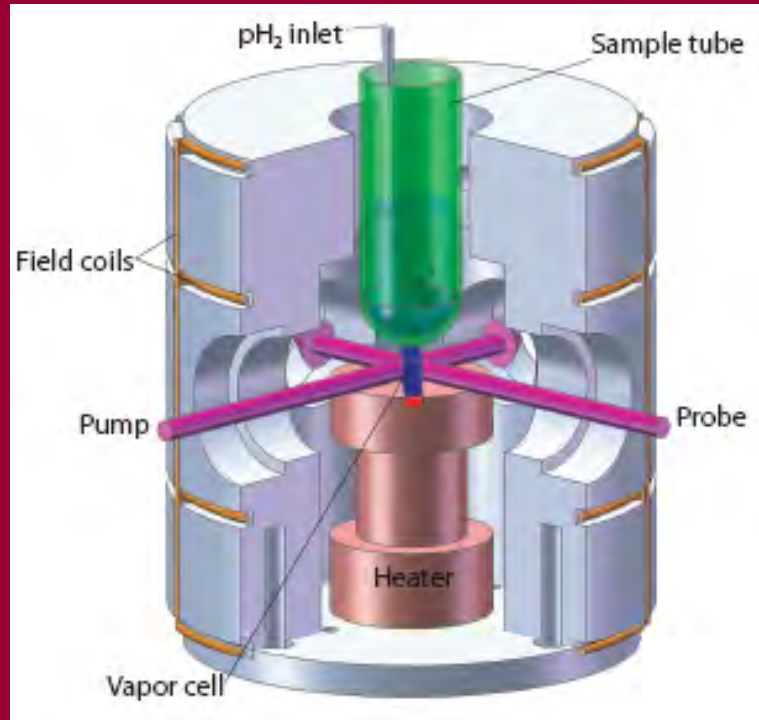


Magnetic  
shields



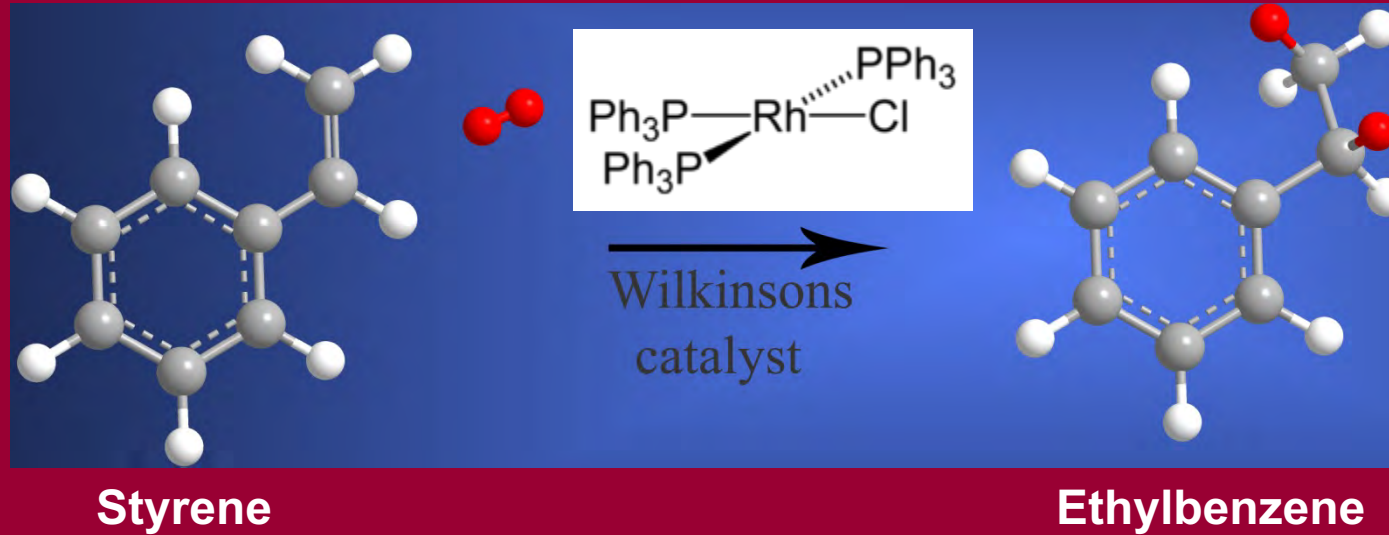
T. Theis  
P. Ganssle  
G. Kervern  
M. P. Ledbetter  
D. B.  
A. Pines

# NMR inside-out: $p\text{H}_2$ polarization; laser-mag detection



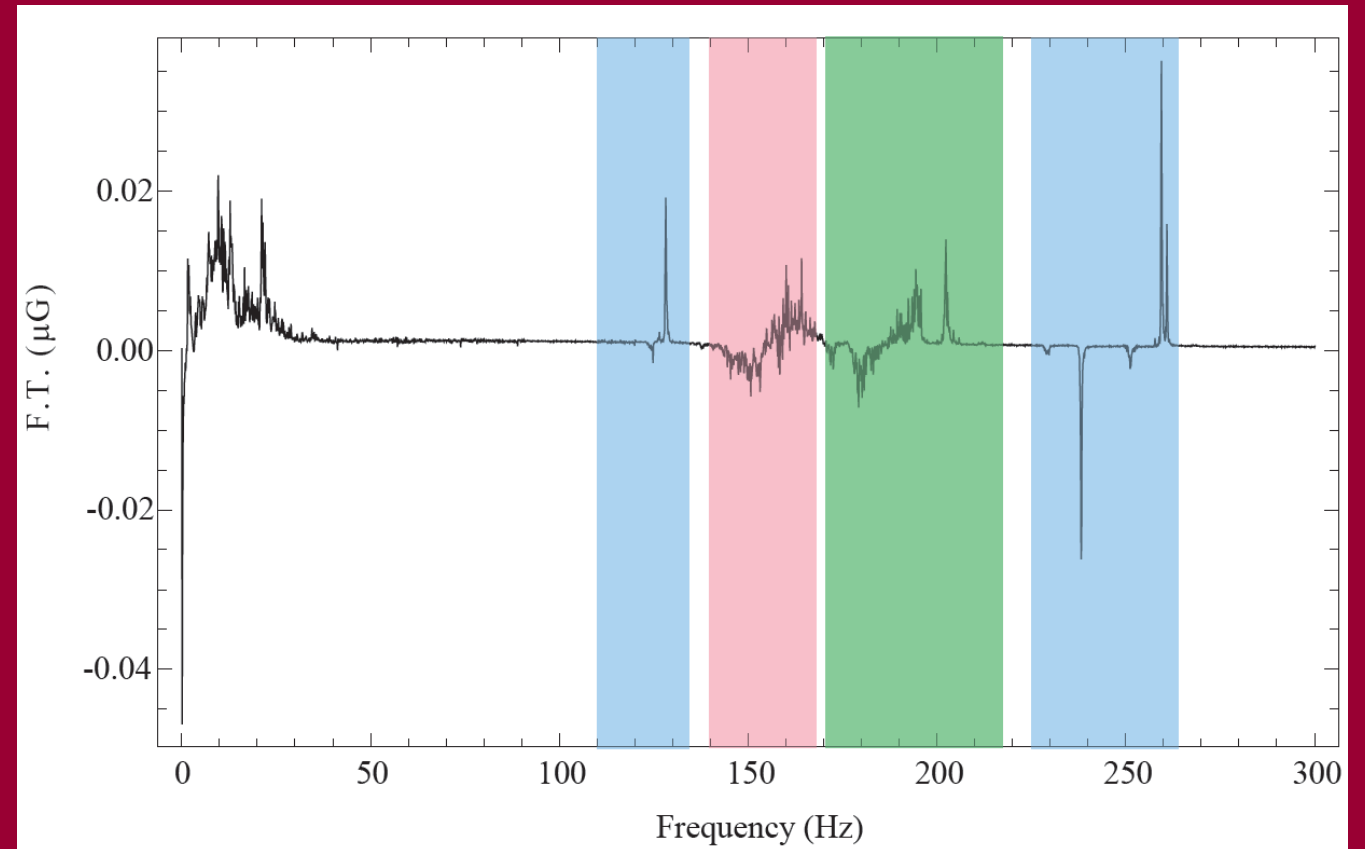
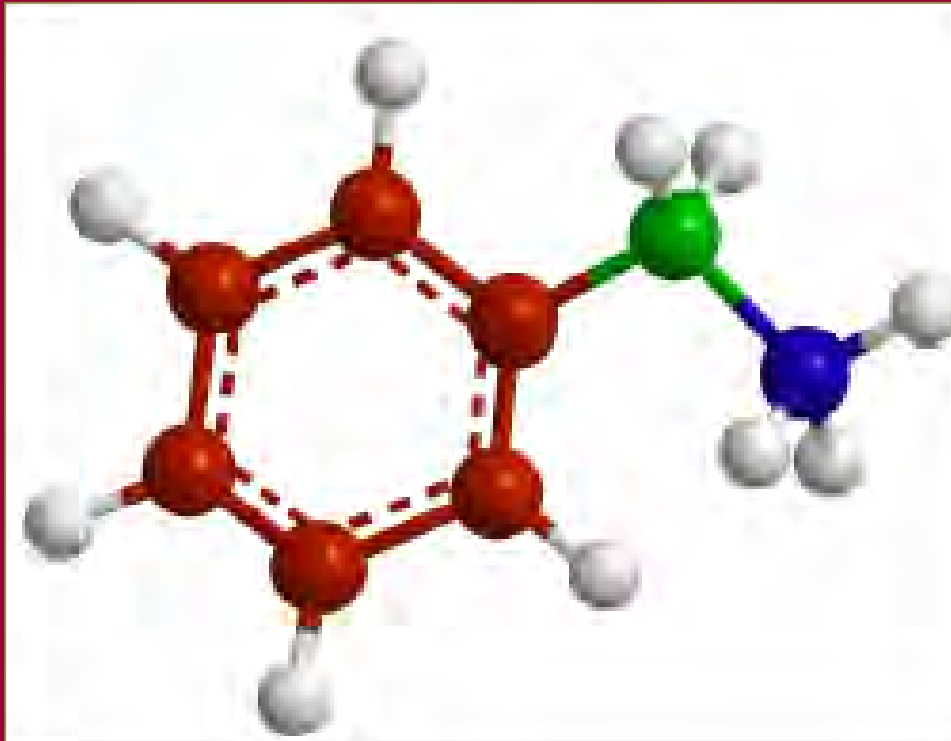


# Hydrogenation with $\text{pH}_2$



# Hydrogenation with $pH_2$

Natural Abundance  
1.1% of  $^{13}C$



# Parahydrogen-enhanced zero-field nuclear magnetic resonance

T. Theis<sup>1,2</sup>, P. Ganssle<sup>1,2</sup>, G. Kervern<sup>1,2</sup>, S. Knappe<sup>3</sup>, J. Kitching<sup>3</sup>, M. P. Ledbetter<sup>4</sup>, D. Budker<sup>4,5</sup> and A. Pines<sup>1,2\*</sup>

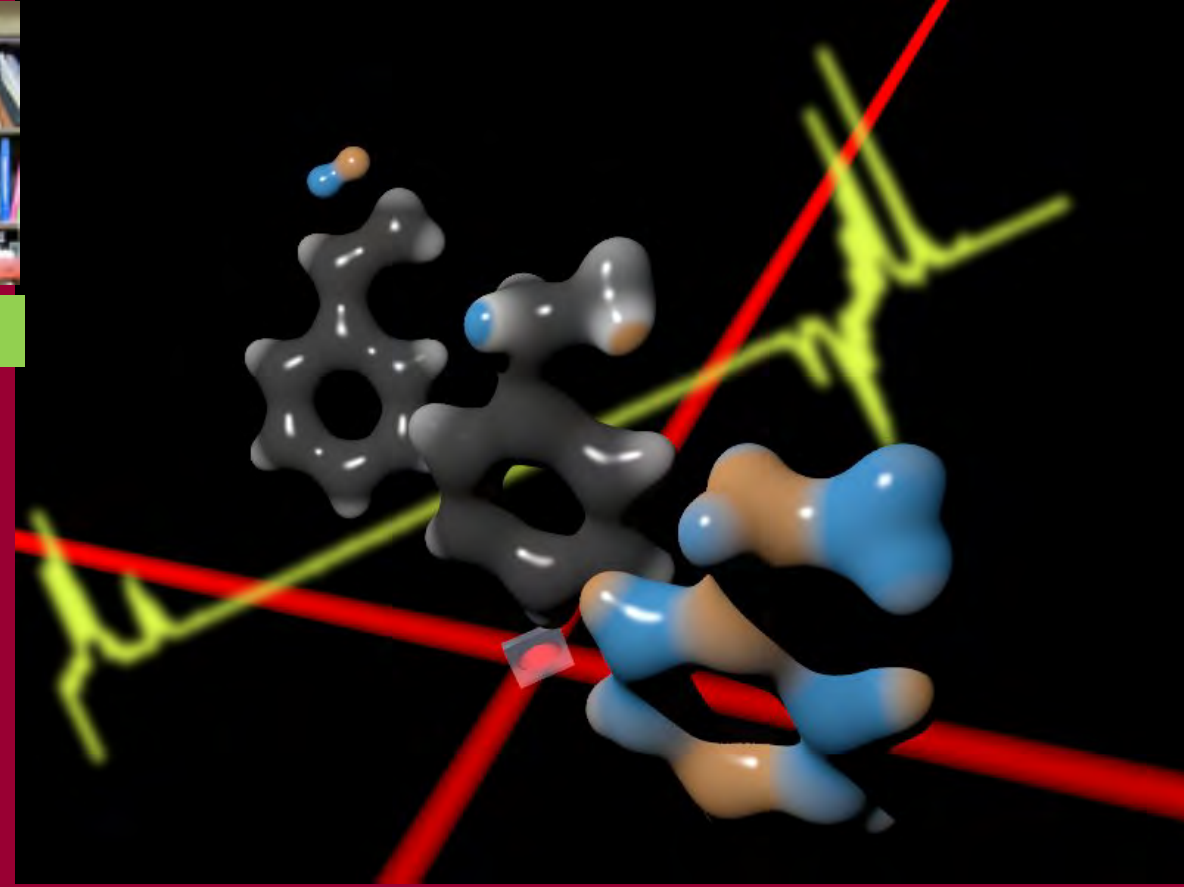


Thomas Theis



Alex Pines

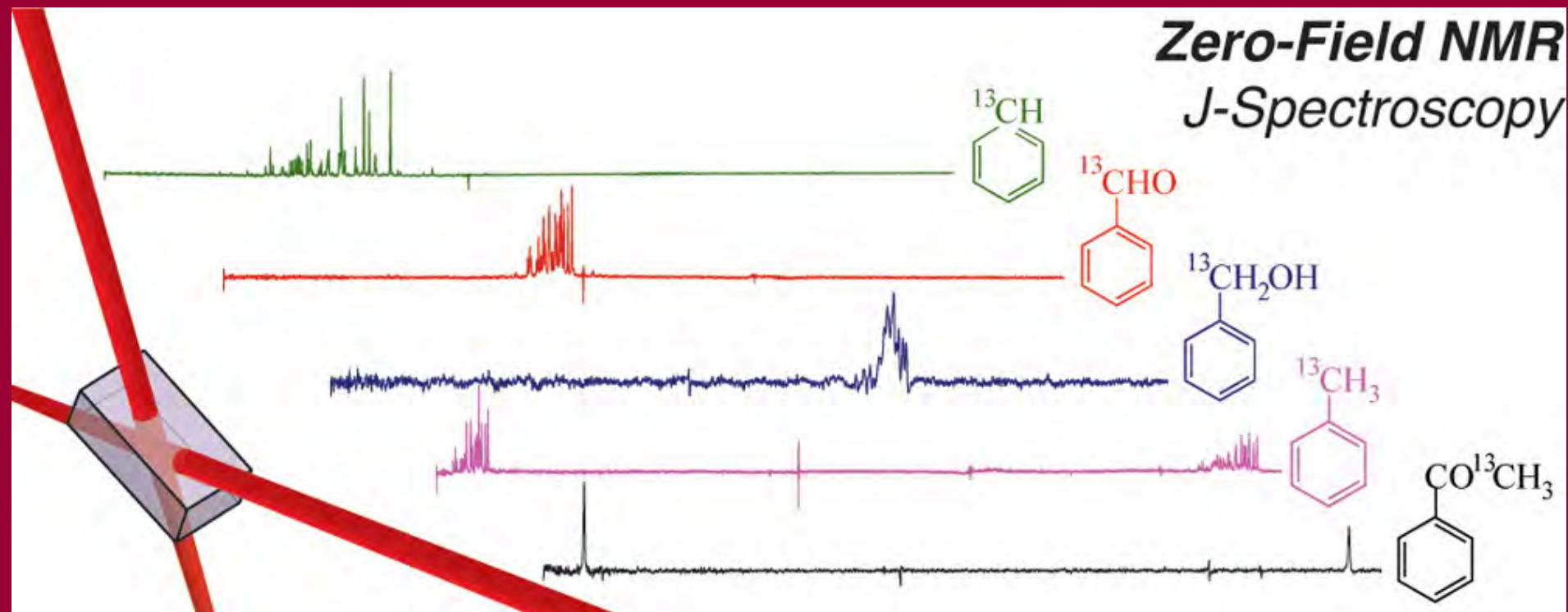
**NMR  
without any  
magnets!**

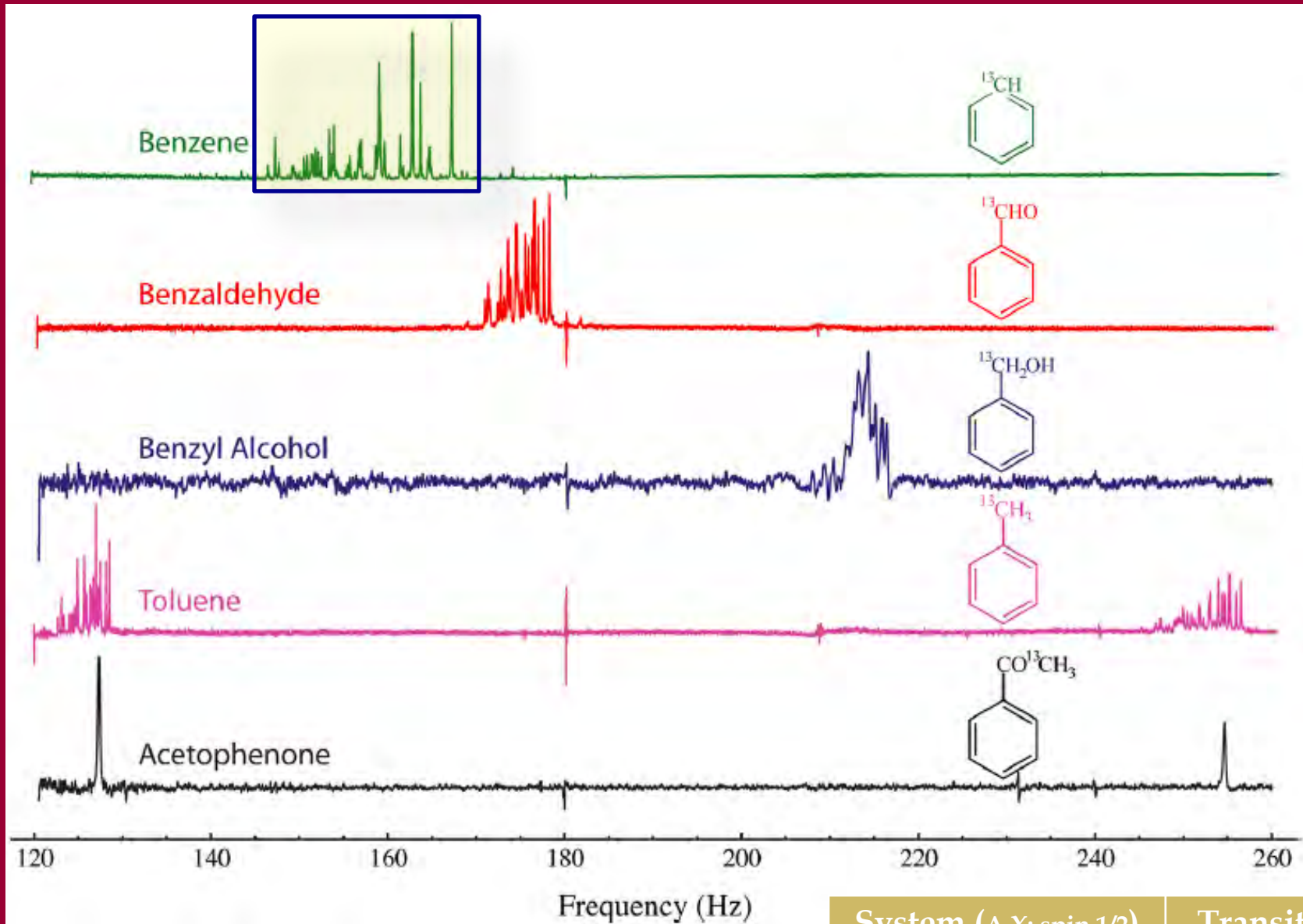




## High-Resolution Zero-Field NMR *J*-Spectroscopy of Aromatic Compounds

John W. Blanchard,<sup>\*,†,‡</sup> Micah P. Ledbetter,<sup>||</sup> Thomas Theis,<sup>†,‡,⊥</sup> Mark C. Butler,<sup>†,‡,#</sup> Dmitry Budker,<sup>§,||</sup> and Alexander Pines<sup>\*,†,‡</sup>





- $\sim 100 \mu\text{l}$  samples
- First-order line positions are **well resolved**
- Line structure reports **higher-order couplings**
- 11 mHz linewidth (HWHM): **high resolution**

| System (A,X: spin 1/2) | Transitions @ |
|------------------------|---------------|
| AX                     | J             |
| AX <sub>2</sub>        | 3J/2          |

# High-Resolution Zero-Field NMR *J*-Spectroscopy of Aromatic Compounds

John W. Blanchard,<sup>\*,†,‡</sup> Micah P. Ledbetter,<sup>||</sup> Thomas Theis,<sup>†,‡,⊥</sup> Mark C. Butler,<sup>†,‡,#</sup> Dmitry Budker,<sup>§,||</sup> and Alexander Pines<sup>\*,†,‡</sup>

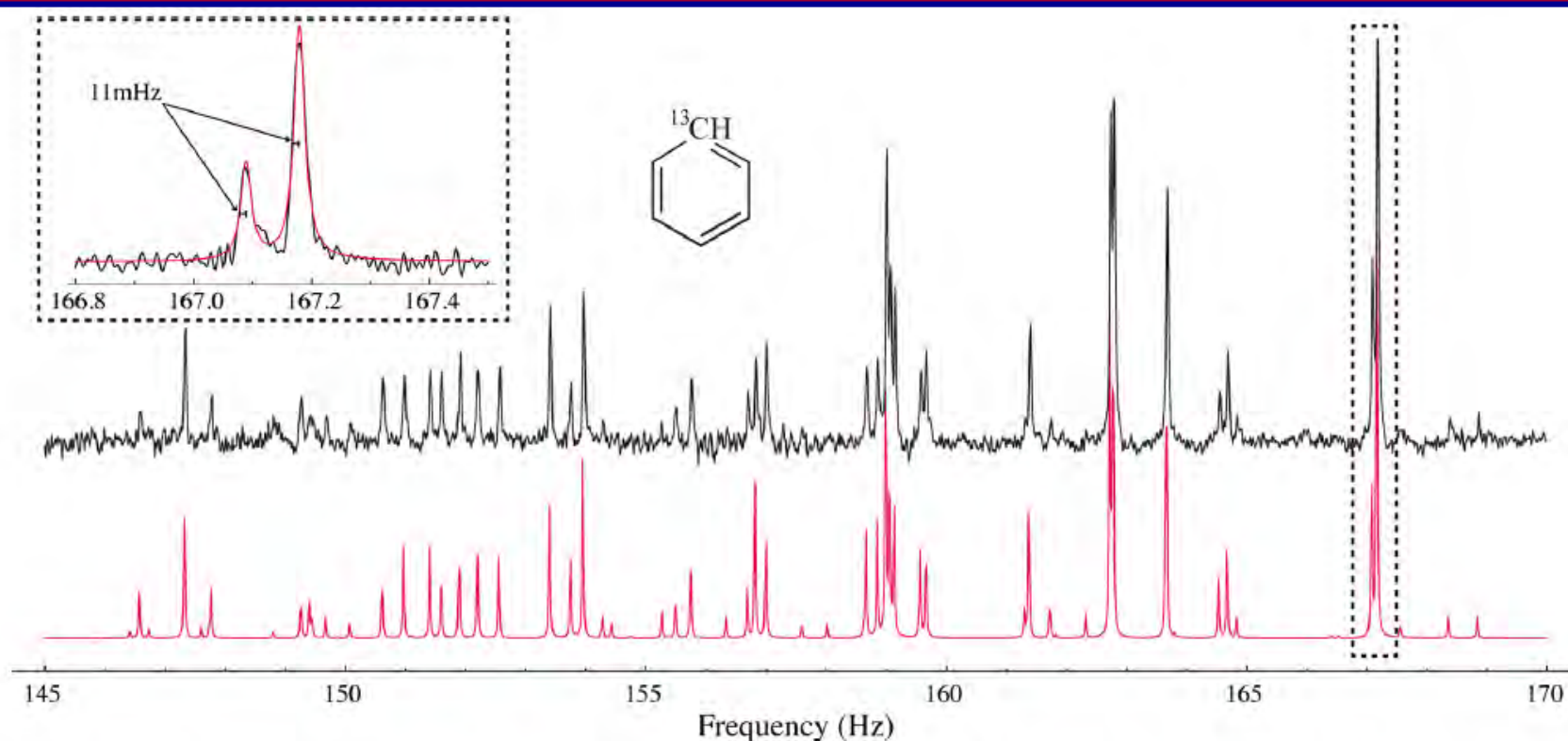
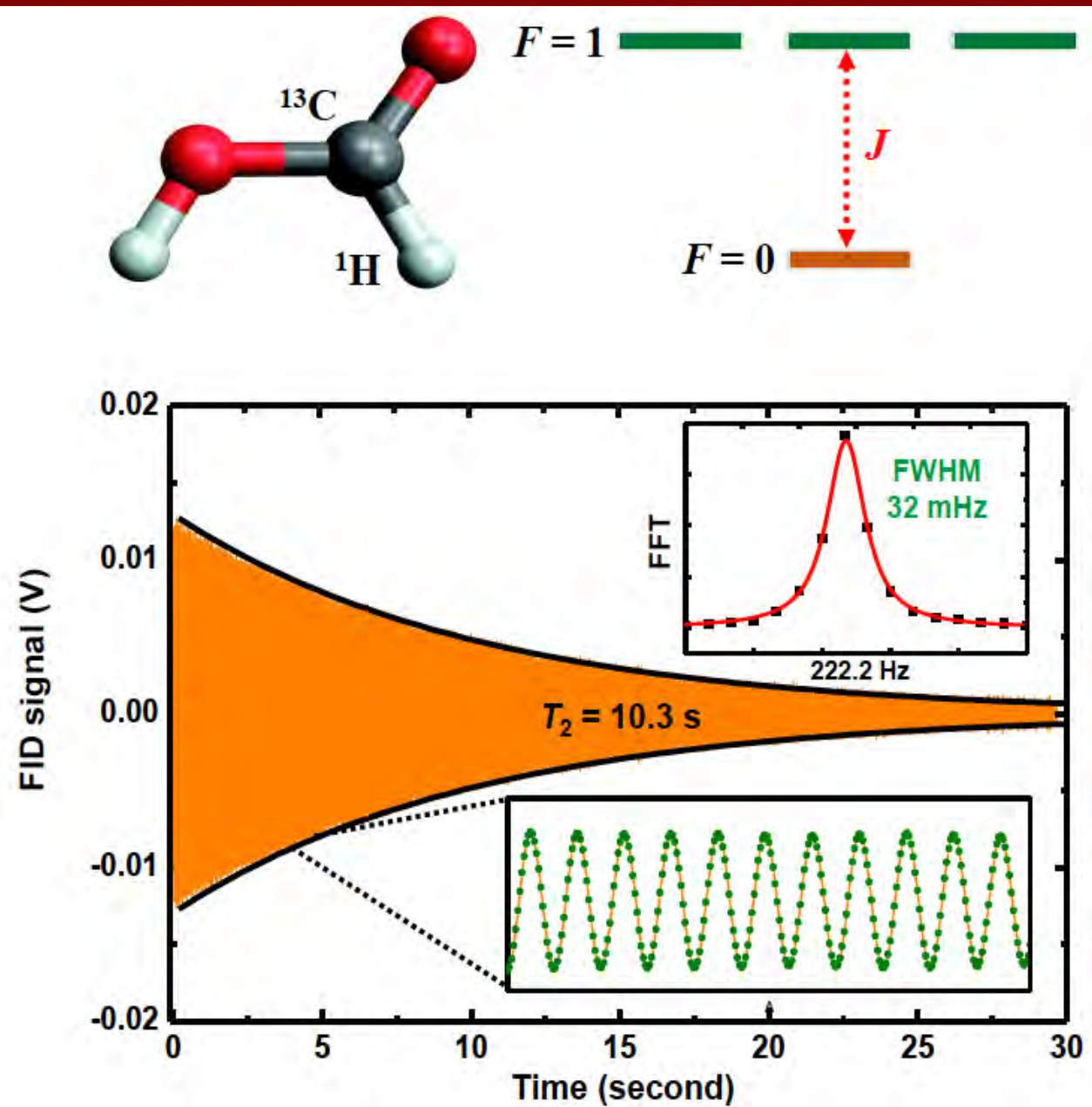
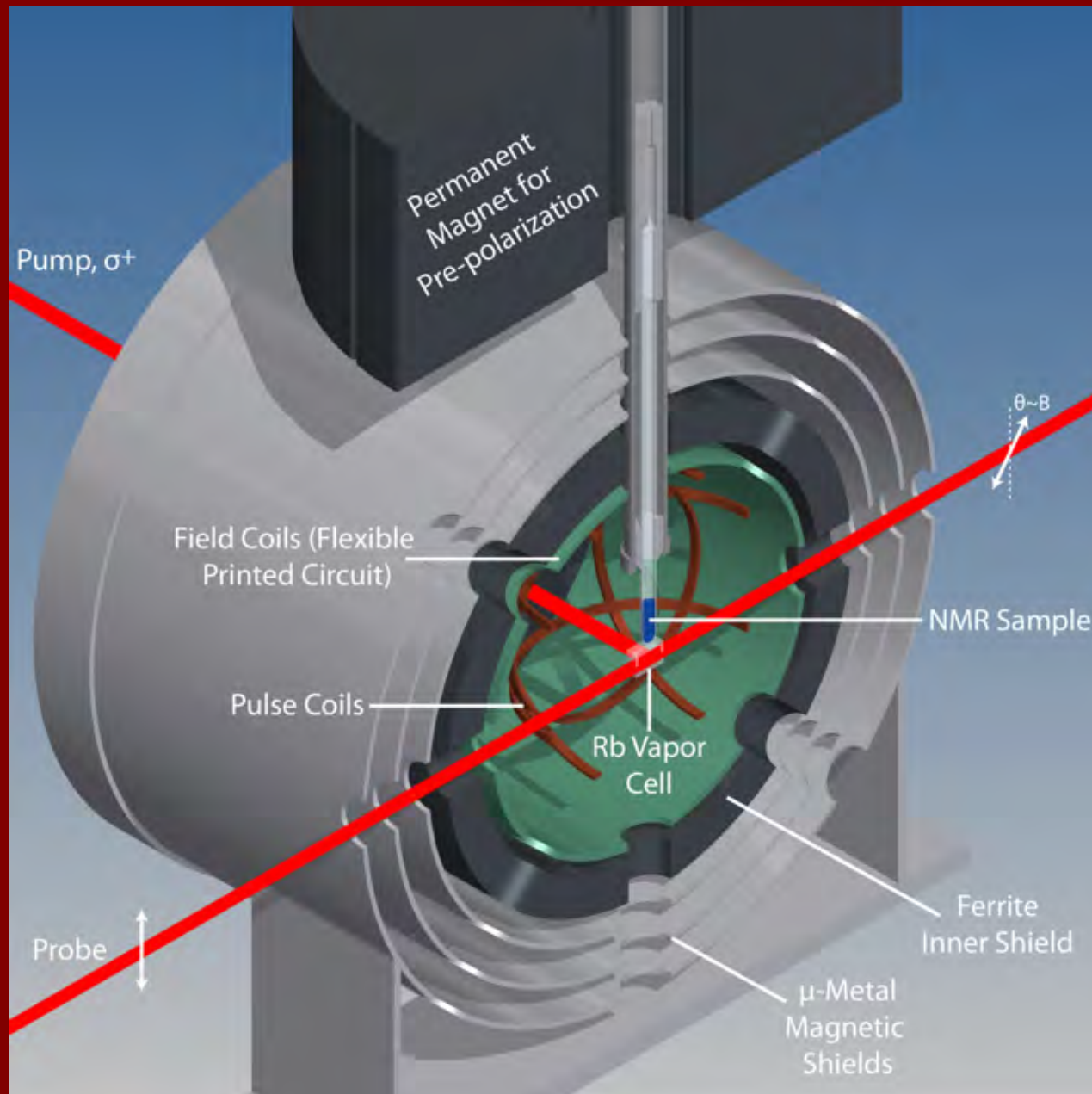
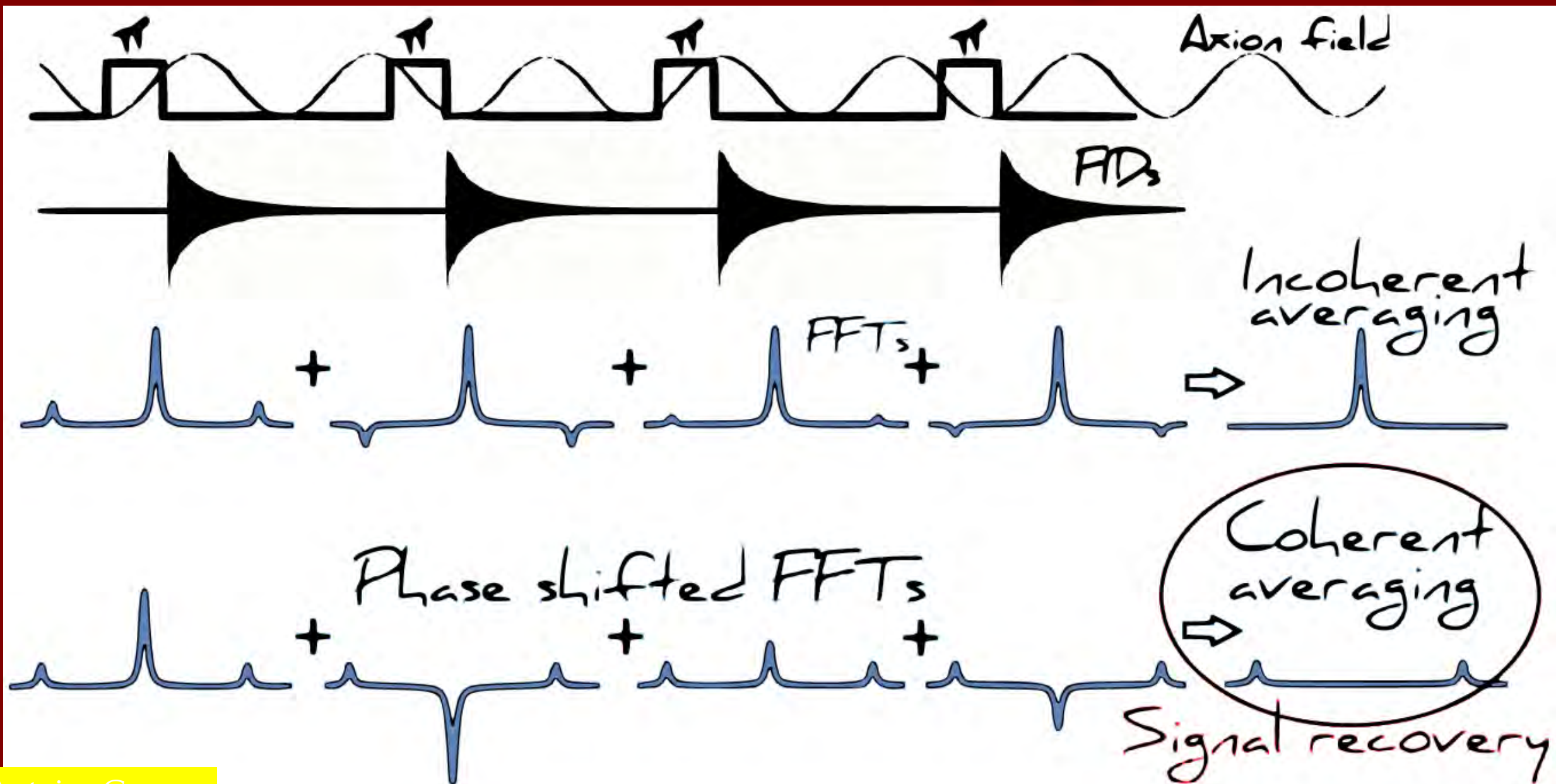


Figure 2. Experimental (upper trace) and simulated (lower trace) spectrum of benzene-<sup>13</sup>C<sub>1</sub> in the neighborhood of <sup>1</sup>J<sub>CH</sub>. Inset shows fitting of two high-frequency peaks with 11 mHz half-width at half-maximum, consistent with Fourier resolution limited by 80s acquisition time.

# CASPER-NOW with ZULF NMR

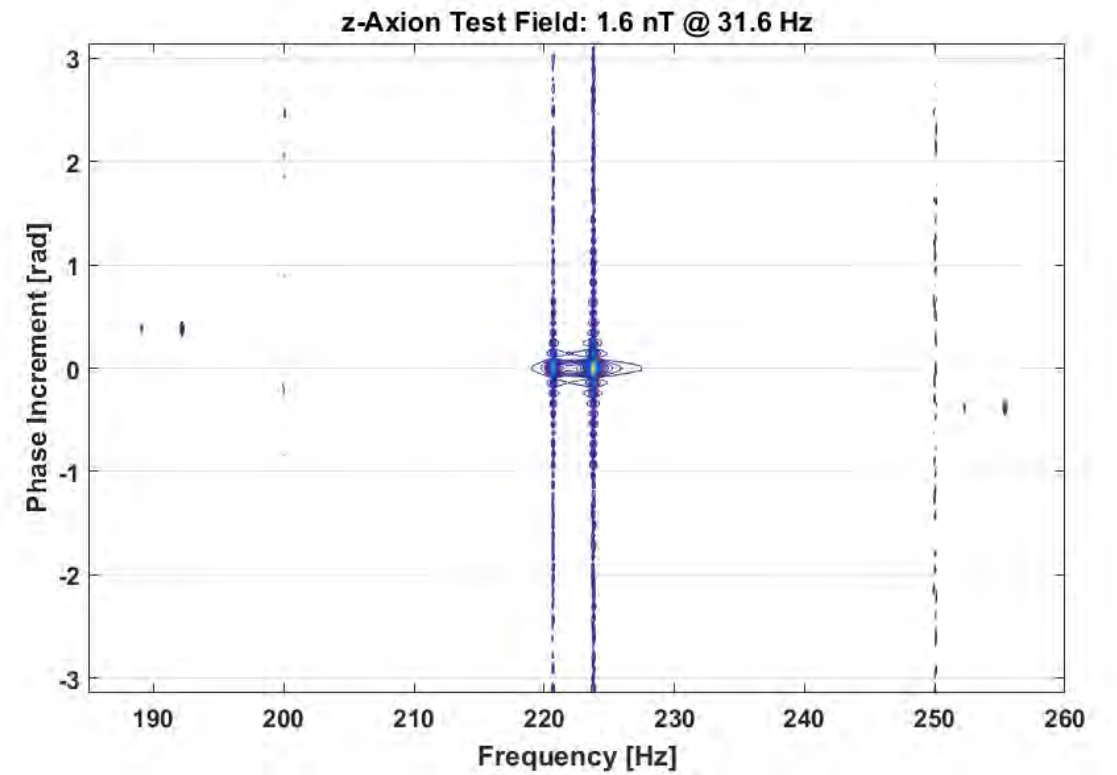
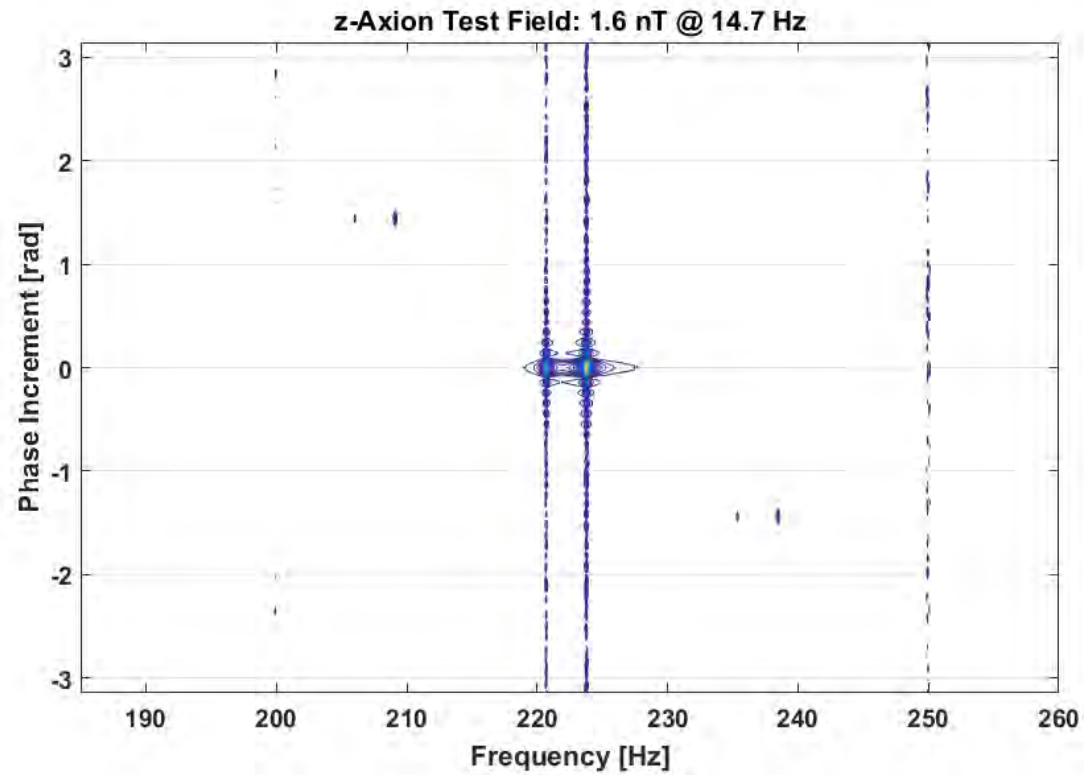


# Coherent Averaging



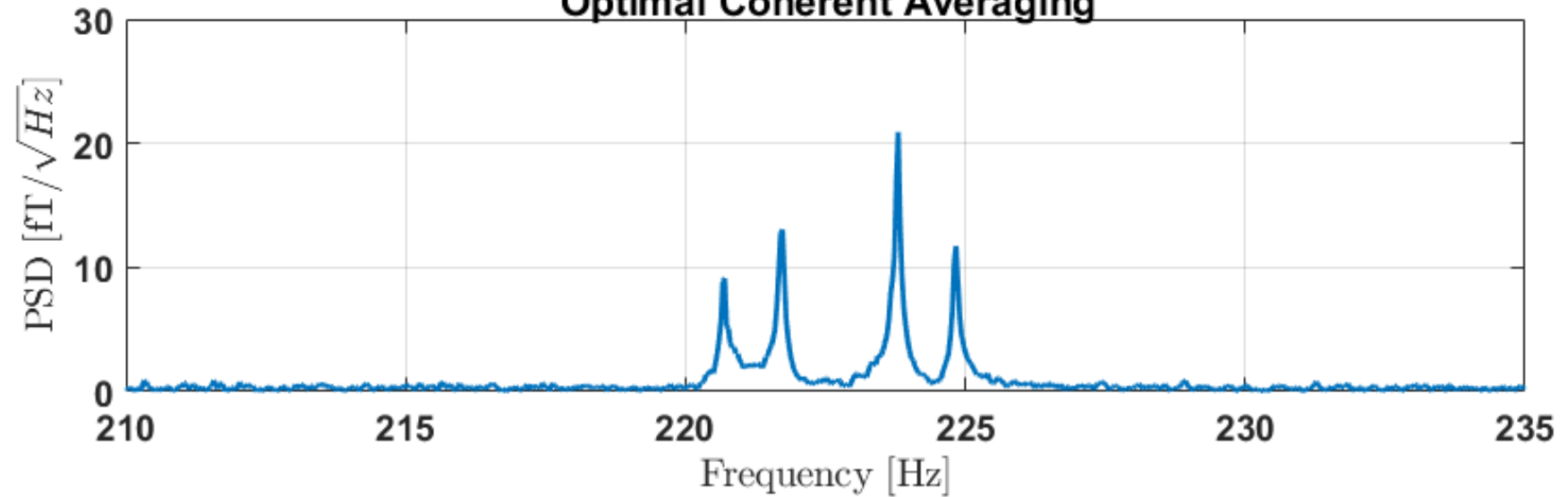


# Coherent Averaging

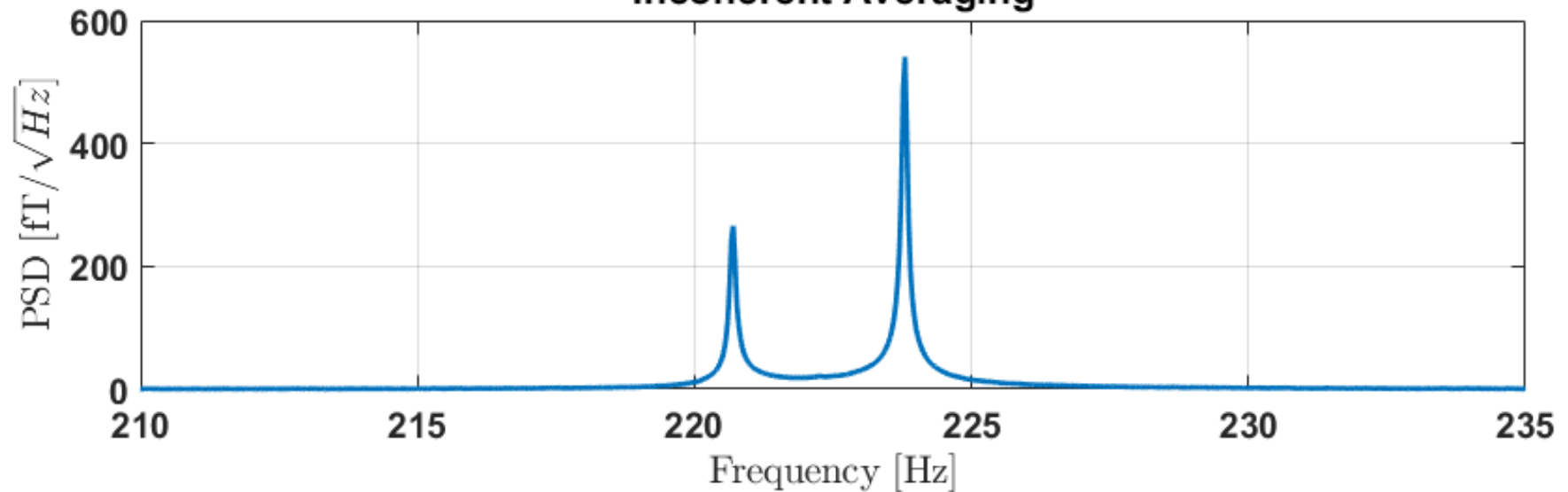


# Coherent Averaging

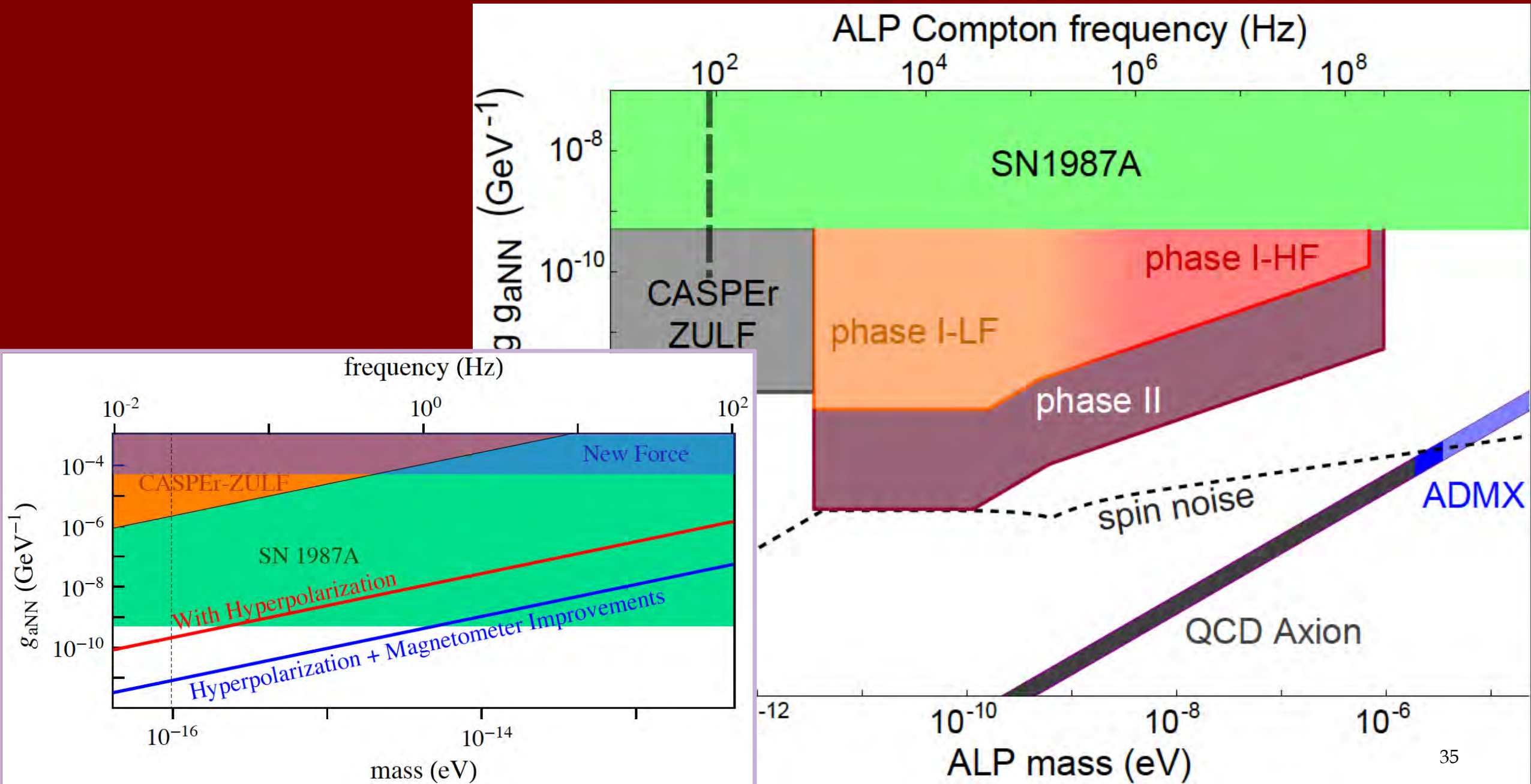
z-Axion TestField 1.6nT @ 1.0 Hz  
Optimal Coherent Averaging



Incoherent Averaging



# CASPER-Wind/NOW



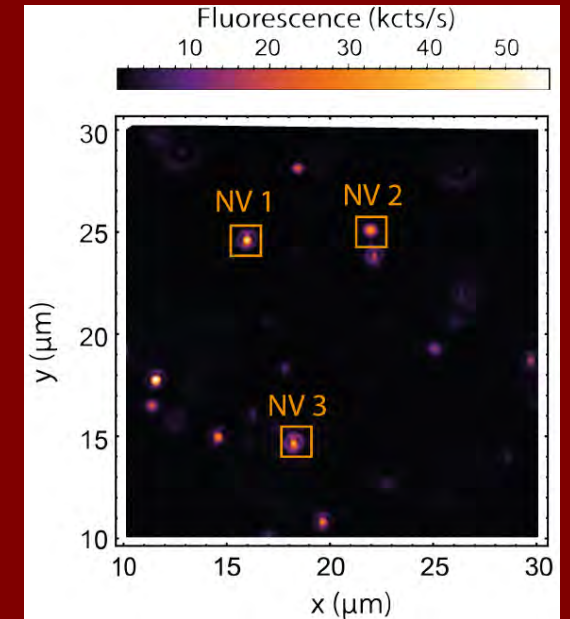
# Summary: fundamental physics with weird NMR

## ✧ Cosmic Axion Spin Precession Experiment

➔ CASPER-E

➔ CASPER-Wind/ZULF/Now

New



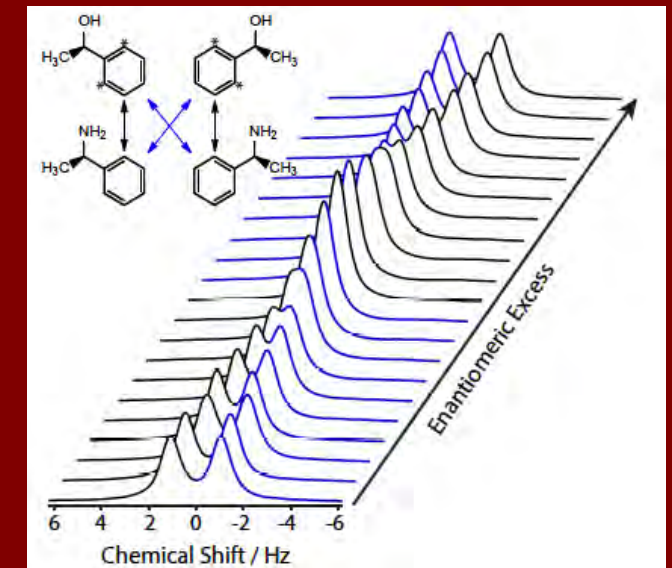
## ✧ Zero- and Ultralow-Field NMR

➔ ParaHydrogen Induced Polarization

➔ J-coupling spectroscopy @ ZULF

➔ NV-ZULF NMR

New



## ✧ Chiral parity violation in NMR

New



Thanks!



European Research Council

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from anywhere in the world

Deutsche  
Forschungsgemeinschaft

DFG



HEISING - SIMONS  
FOUNDATION



HIM

Helmholtz-Institut Mainz

SIMONS FOUNDATION

# At Berkeley



Sean Lourette

Dr. Tao Wang

D.B.

Dr. Andrey Jarmola

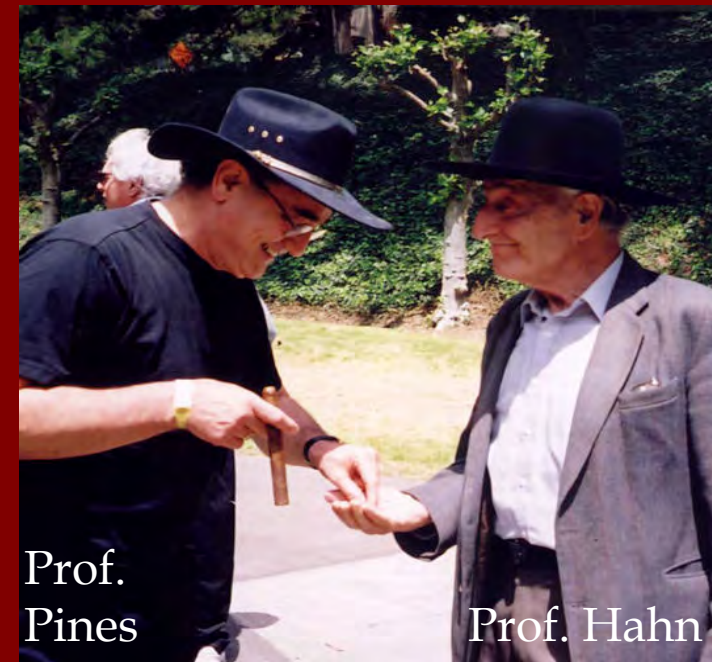
Dr. Metin Kayci



Dr. Vincent Dumont



Michael Solarz

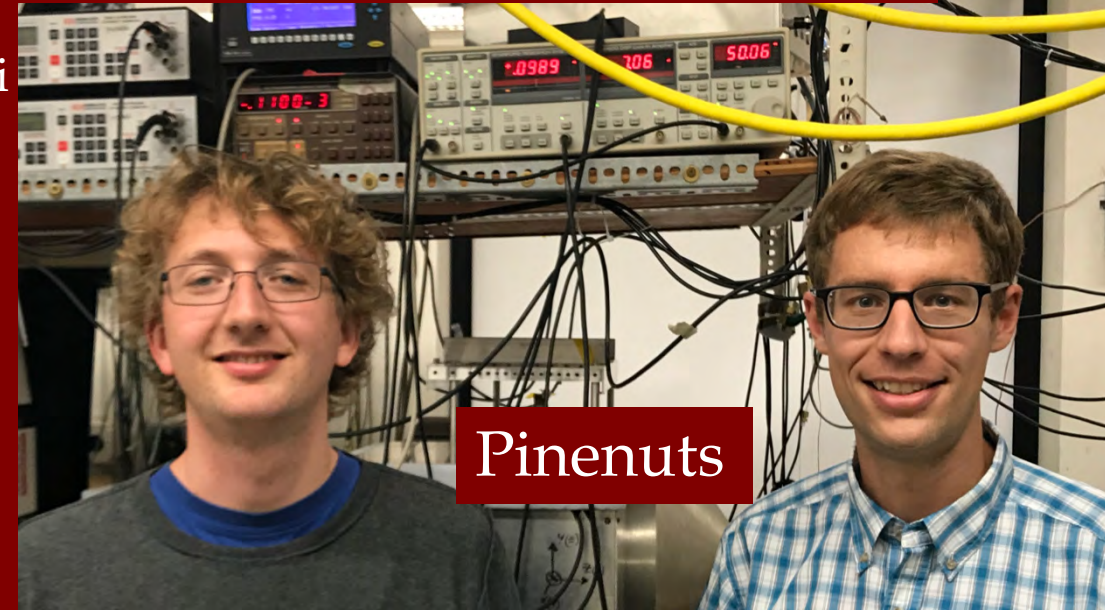


Prof. Pines

Prof. Hahn



SIMONS FOUNDATION



Pinenuts

Tobias Sjolander

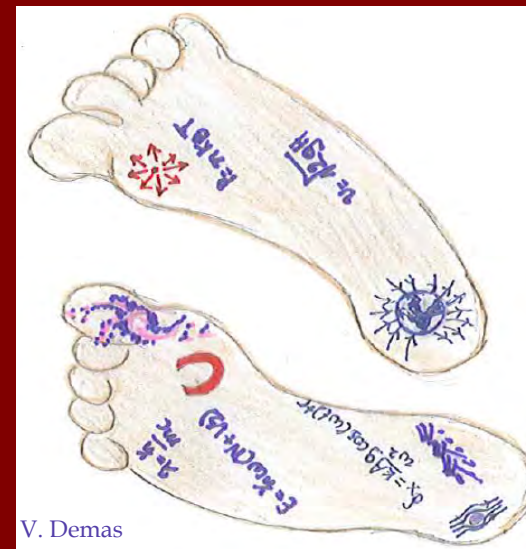
Dr. Jonathan King

# Suggested Questions

- ▣ So what about **parity violation in chiral molecules?**
- ▣ What was that **Maxwell-Proca galactic business?**
- ▣ What are some other ways to **search for DM axions?** (**GNOME [video](#)**)
- ▣ Tell us more about **single-spin NMR**
- ▣ What is the latest in **atomic and diamond magnetometers?**
- ▣ Can you do **magnetic resonance 100 km up in the sky?**
- ▣ What is **Physics on Your Feet?**

# Suggested Questions

1. So what about **parity violation in chiral molecules**?
2. What was that **Maxwell-Proca galactic business**?
3. What are some other ways to **search for DM axions**? ([GNOME video](#))
4. Tell us more about **single-spin NMR**
5. What is the latest in **atomic and diamond magnetometers**?
6. Can you do **magnetic resonance 100 km up in the sky**?
7. What is **Physics on Your Feet**?

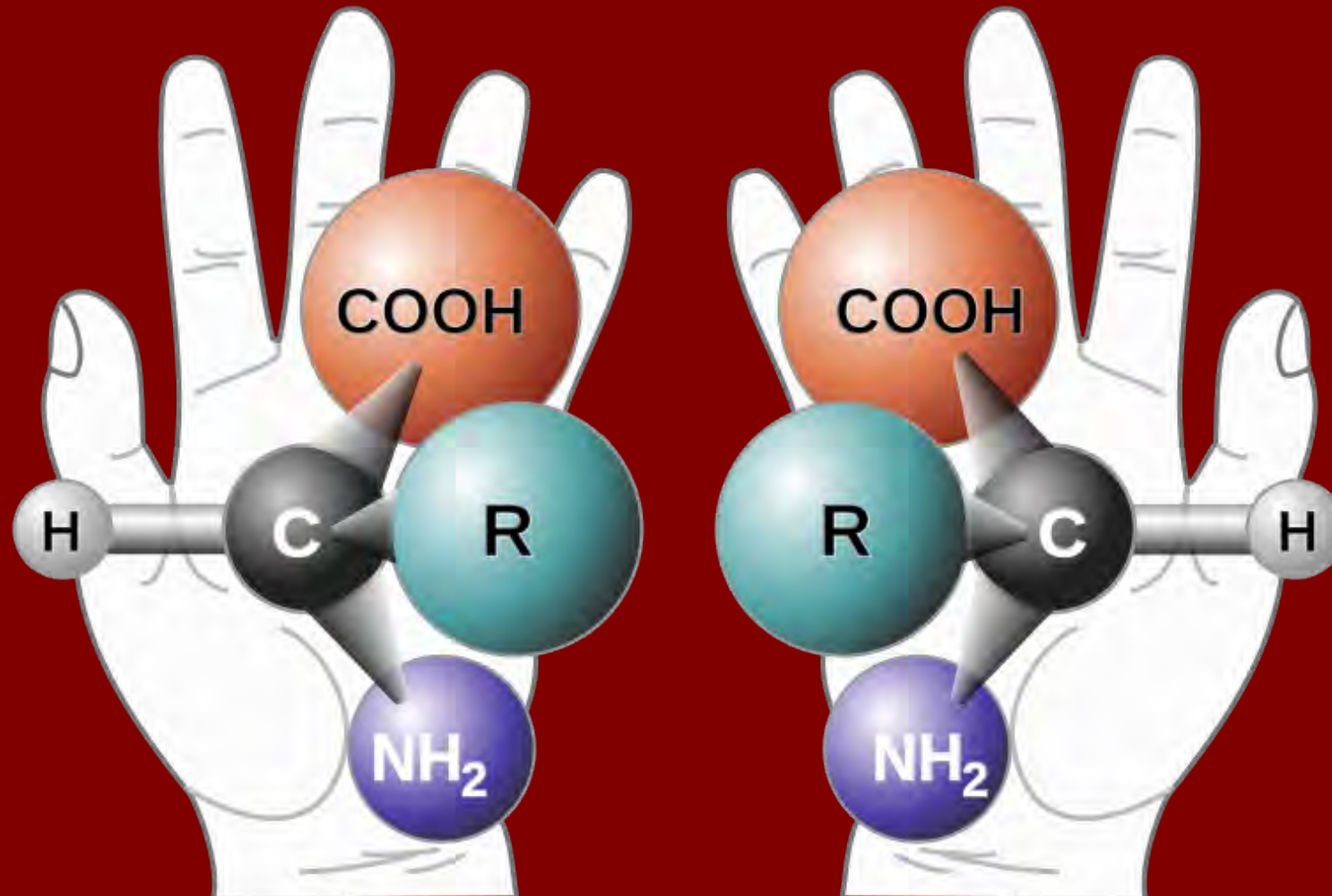




# Measuring molecular parity non-conservation using NMR Spectroscopy

J. Eills,<sup>1,2</sup> J. W. Blanchard,<sup>3,4,5</sup> L. Bougas,<sup>2,6</sup> M. G. Kozlov,<sup>7</sup> A. Pines,<sup>5,4</sup> and D. Budker<sup>2,3,6,8</sup>

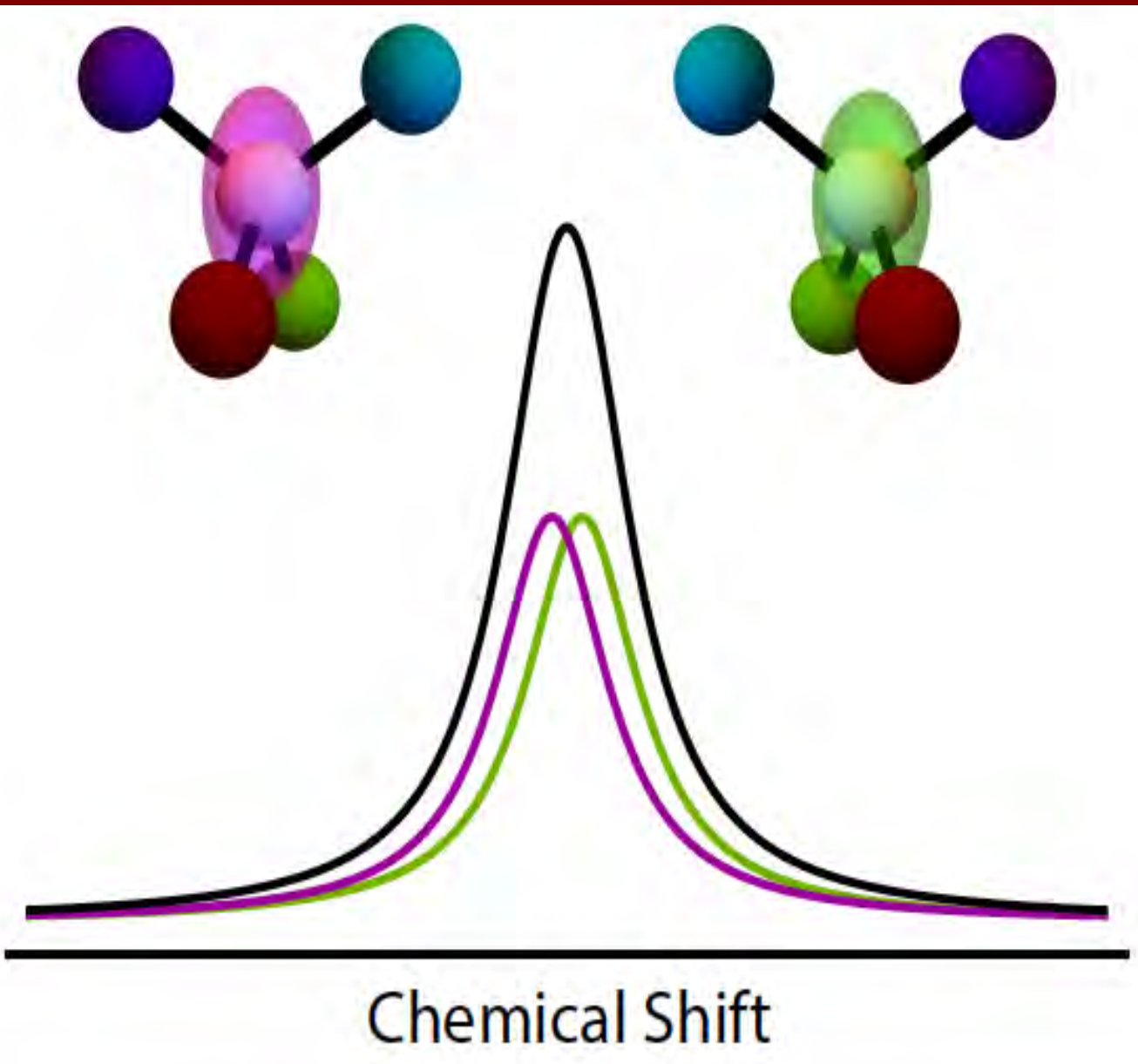
[Phys. Rev. A 96, 042119](#) – Published 30 October 2017



# A PROVOCATIVE QUESTION:

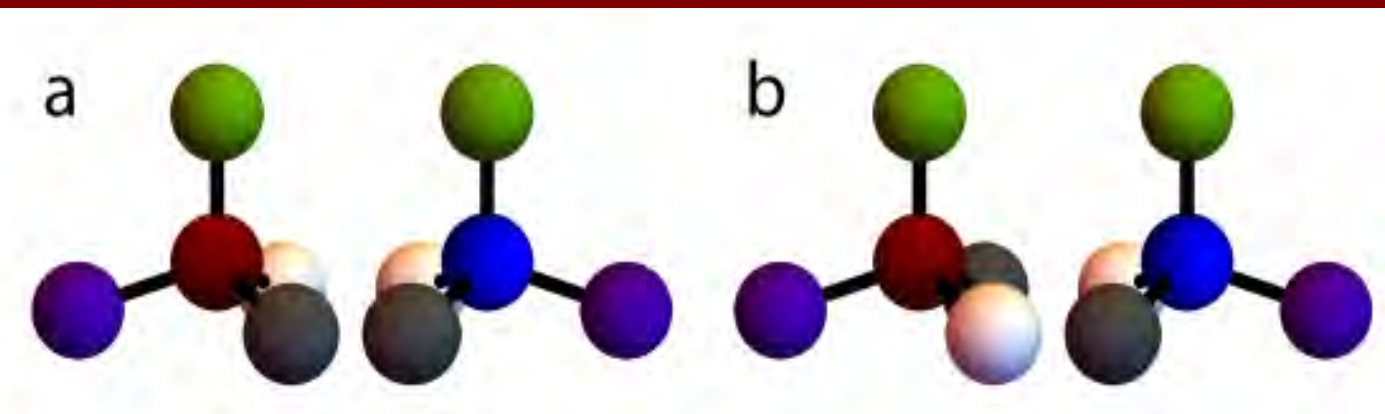
Why do **chiral** molecules have  
first-order PNC energy shifts ?  
(While this is normally forbidden)

# PNC in racemic mixtures: impossible?

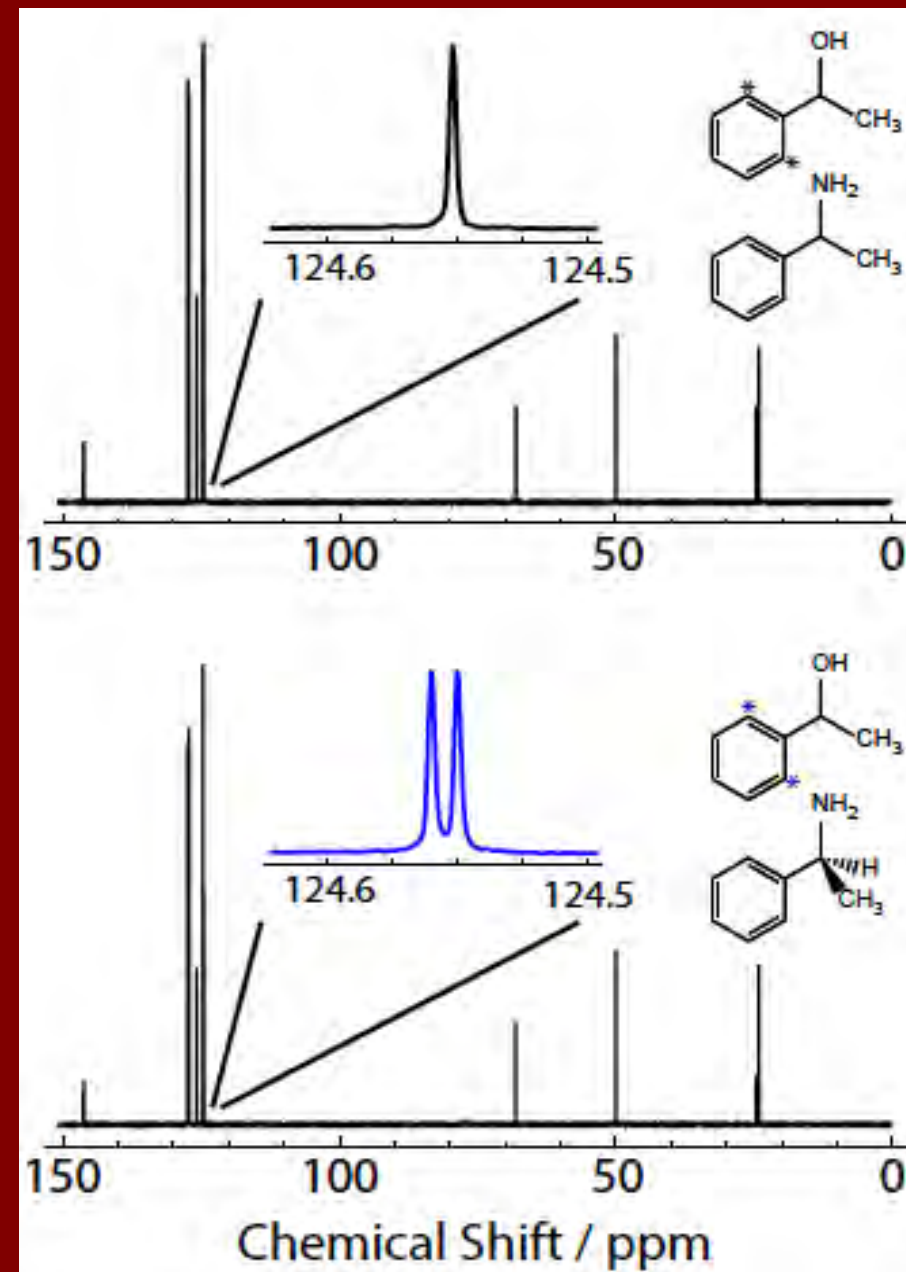


- PNC in chiral mol.  $\rightarrow$  energy shift
- PNC in NMR  $\rightarrow$  Nucl.Spin.Dep PNC
- $B=20\text{ T} \rightarrow \sim 1\text{ mHz}$  line shifts
- No way in a mixture...

# Diastereomeric NMR shift



James Eills (SOTON)  
John Blanchard  
Lykourgos Bougas  
Mikhail Kozlov  
Alexander Pines  
D.B.



# Diastereomeric NMR shift

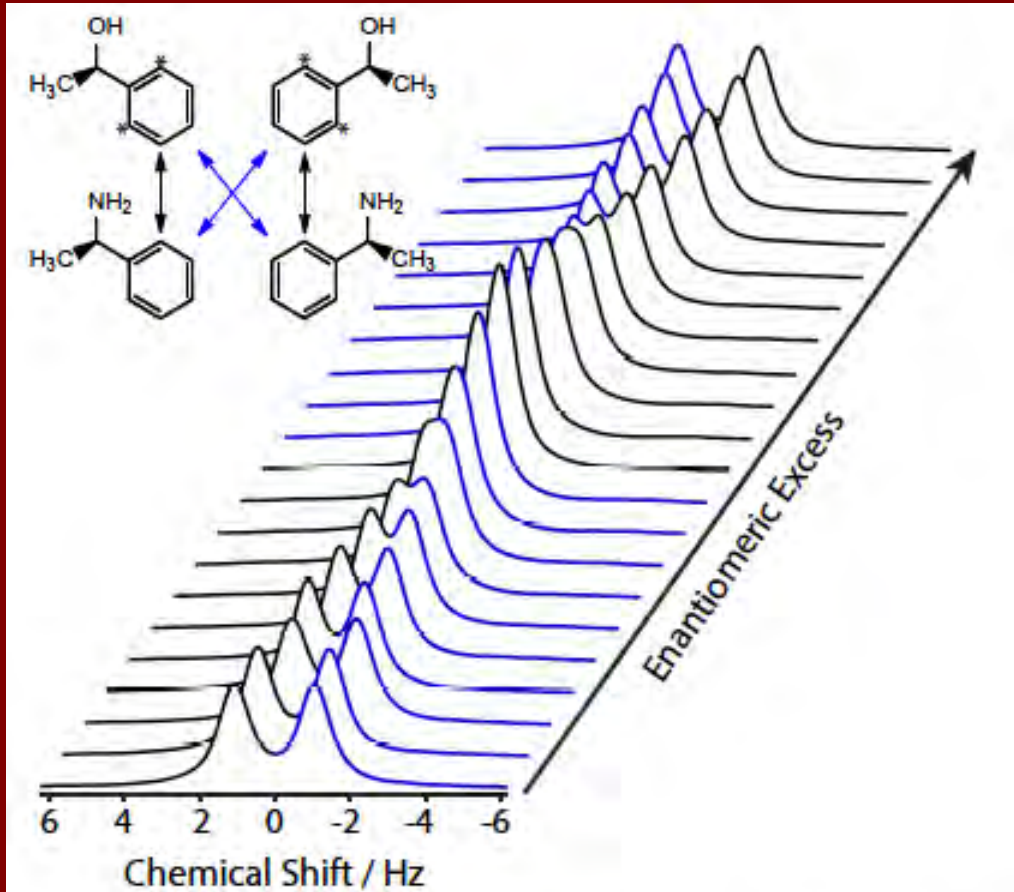


FIG. 4. Stacked  $^{13}\text{C}$  spectra showing diastereomeric splitting of 1-phenylethanol as the enantiomeric excess of the 1-phenylethylamine environment is varied. The scale is in hertz, and centered on the peak of interest. All spectra were acquired at 298 K by averaging 32 transients, with proton decoupling, and have line broadening [35] of 0.5 Hz applied. The inset shows the four possible diastereomeric interactions between the sensor (1-phenylethanol) and chiral solvating reagent (1-phenylethylamine).

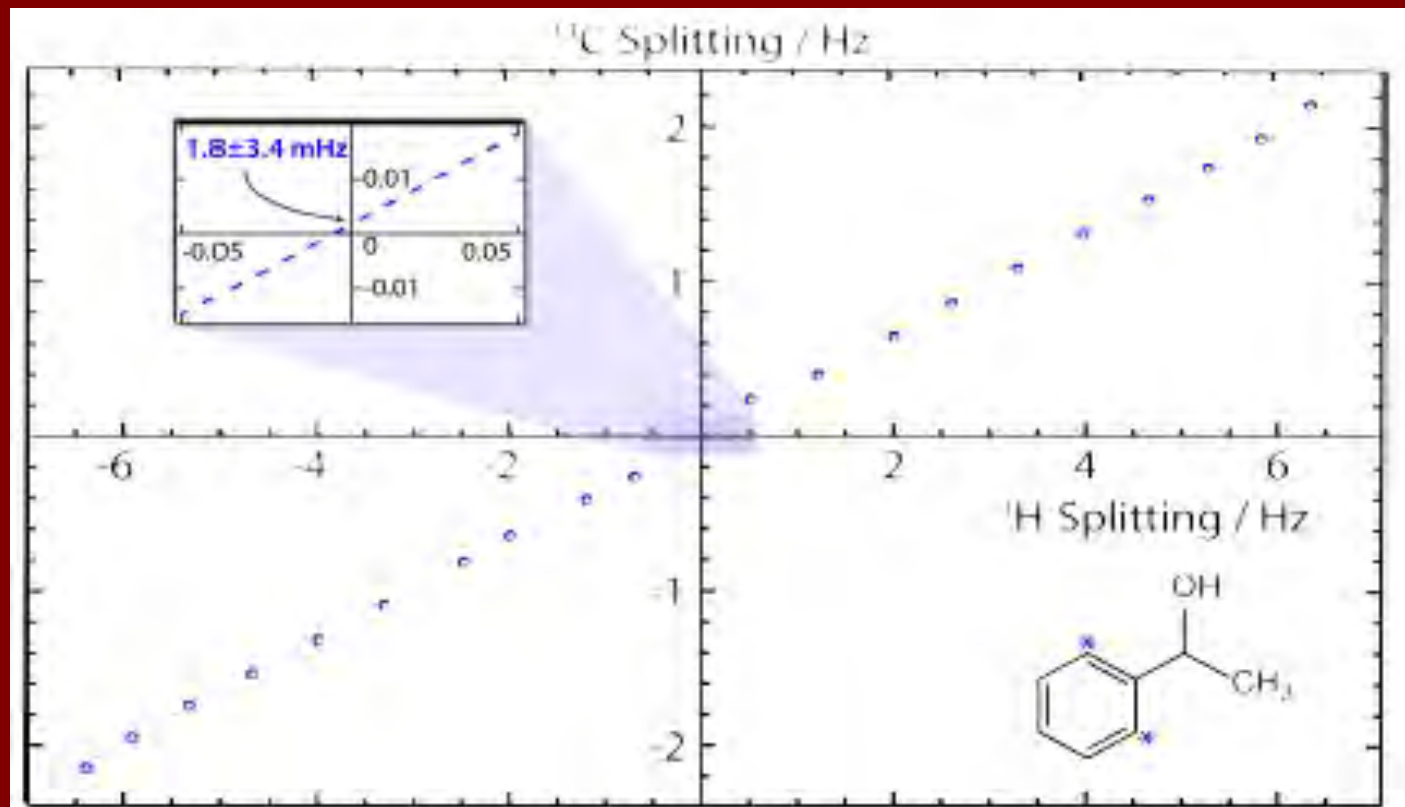


FIG. 5. Experimental data showing the diastereomeric splitting of the  $^{13}\text{C}$  peaks as a function of the 1-phenylethylamine enantiomeric excess. Data points were acquired at 20 T and 298 K, by averaging 32 transients. The enantiomeric excess of each solution was determined by measuring the  $^1\text{H}$  splitting, as discussed in more detail in the text.

“Built-in comagnetometer” !!!

## Bottom line(s):

- Measure chiral PNC w/ **racemic mixtures**
- Built-in  $^1\text{H}$  “**comagnetometer**”
- Systematics seem tractable
- It may, indeed, be **possible to detect chiral PNC in NMR**



**A hypothetical effect  
of  
Maxwell-Proca electromagnetic stresses  
on  
galaxy rotation curves**

**D.D. Ryutov, Dmitry Budker, and V.V. Flambaum**

[arXiv:1708.09514](https://arxiv.org/abs/1708.09514)



**Dmitri Ryutov Wins 2017  
Maxwell Prize for Plasma Physics**



# Finite Photon Mass?

Citation: C. Patrignani *et al.* (Particle Data Group), *Chin. Phys. C*, **40**, 100001 (2016) and 2017 update

$\gamma$  (photon)

$$I(J^{PC}) = 0,1(1^{- -})$$

## $\gamma$ MASS

Results prior to 2008 are critiqued in GOLDHABER 10. All experimental results published prior to 2005 are summarized in detail by TU 05.

The following conversions are useful:  $1 \text{ eV} = 1.783 \times 10^{-33} \text{ g} = 1.957 \times 10^{-6} m_e$ ;  $\lambda_C = (1.973 \times 10^{-7} \text{ m}) \times (1 \text{ eV}/m_\gamma)$ .

| <u>VALUE (eV)</u>   | <u>CL%</u> | <u>DOCUMENT ID</u>          | <u>TECN</u> | <u>COMMENT</u>                |
|---|------------|-----------------------------|-------------|-------------------------------|
| $<1 \times 10^{-18}$  |            | <sup>1</sup> RYUTOV 07      |             | MHD of solar wind             |
| • • • We do not use the following data for averages, fits, limits, etc. • • • |            |                             |             |                               |
| $<1.8 \times 10^{-14}$  |            | <sup>2</sup> BONETTI 16     |             | Fast Radio Bursts, FRB 150418 |
| $<1.9 \times 10^{-15}$  |            | <sup>3</sup> RETINO 16      |             | Ampere's Law in solar wind    |
| $<2.3 \times 10^{-9}$   | 95         | <sup>4</sup> EGOROV 14      | COSM        | Lensed quasar position        |
|   |            | <sup>5</sup> ACCIOLY 10     |             | Anomalous magn. mom.          |
| $<1 \times 10^{-26}$  |            | <sup>6</sup> ADELBERGER 07A |             | Proca galactic field          |
| no limit feasible   |            | <sup>6</sup> ADELBERGER 07A |             | $\gamma$ as Higgs particle    |

# Effect of Photon Mass on Galaxies?



NGC 4414, a typical spiral galaxy, is about 55,000 light-years in diameter and approximately 60 million light-years away from Earth

## Key points:

- Sufficiently strong forces to explain galactic rotation curves without dark matter
- The effect of mass is indirect, through MHD

# Maxwell-Proca Quasi-Static Electrodynamics



NGC 4414, a typical spiral galaxy, is about 55,000 light-years in diameter and approximately 60 million light-years away from Earth

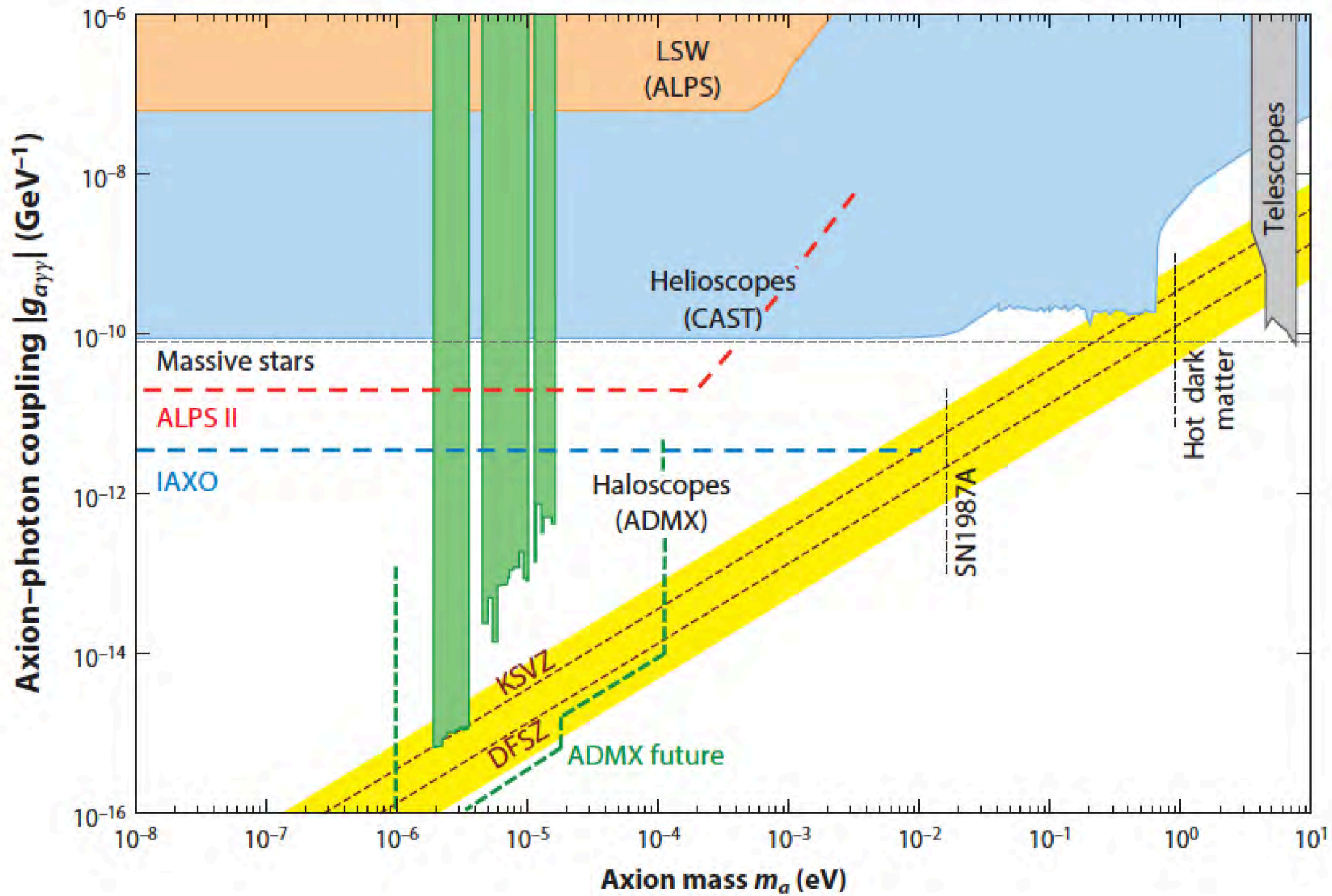
$$\nabla \cdot \mathbf{A} = 0$$

$$\nabla \times \mathbf{E} = -\frac{1}{c} \frac{\partial \mathbf{B}}{\partial t}$$

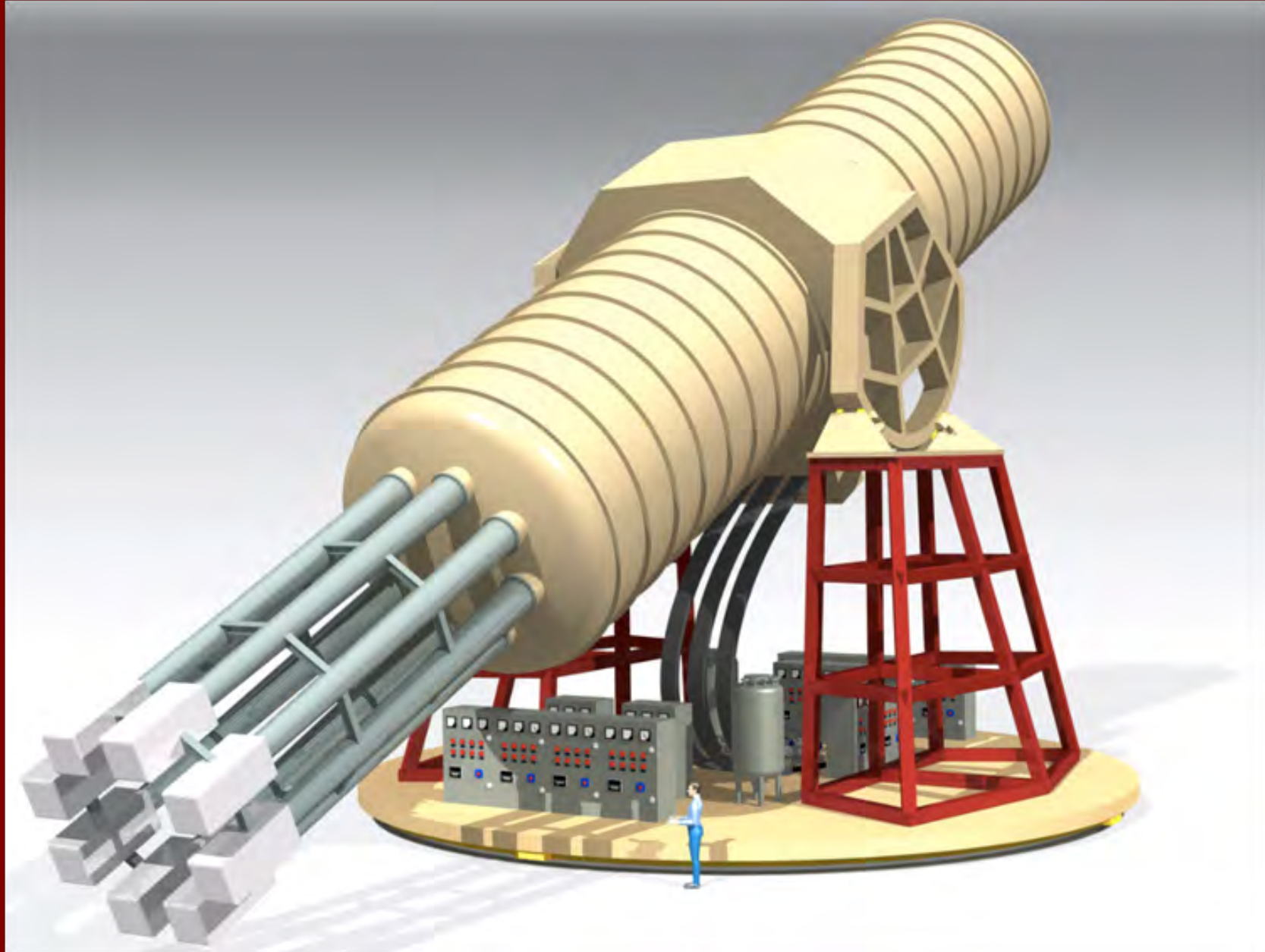
$$\nabla \times \mathbf{A} = \mathbf{B}$$

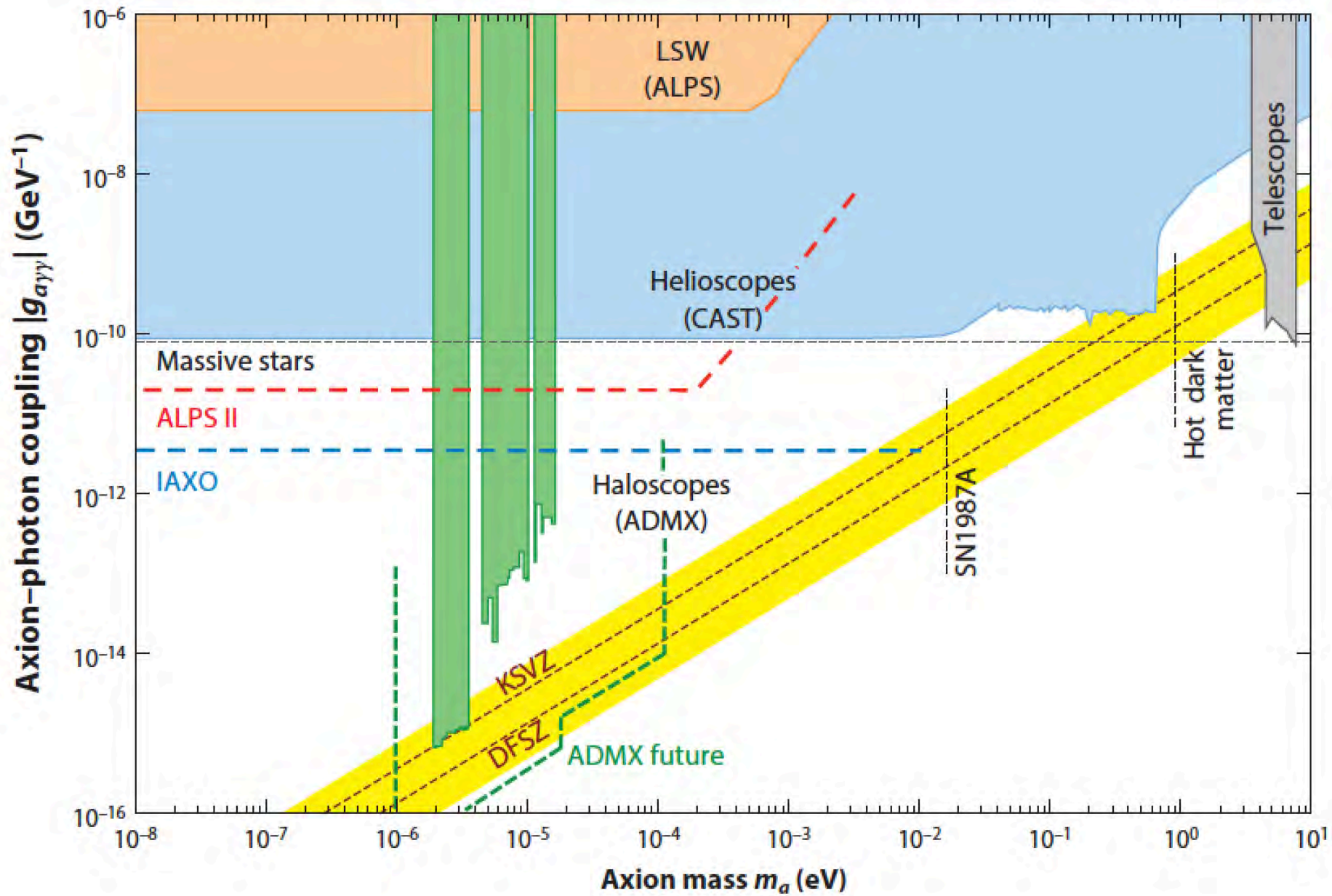
$$\nabla \times \mathbf{B} + \frac{\mathbf{A}}{\hat{\lambda}^2} = \frac{4\pi}{c} \mathbf{j}$$

# A few Axion/ALP experiments

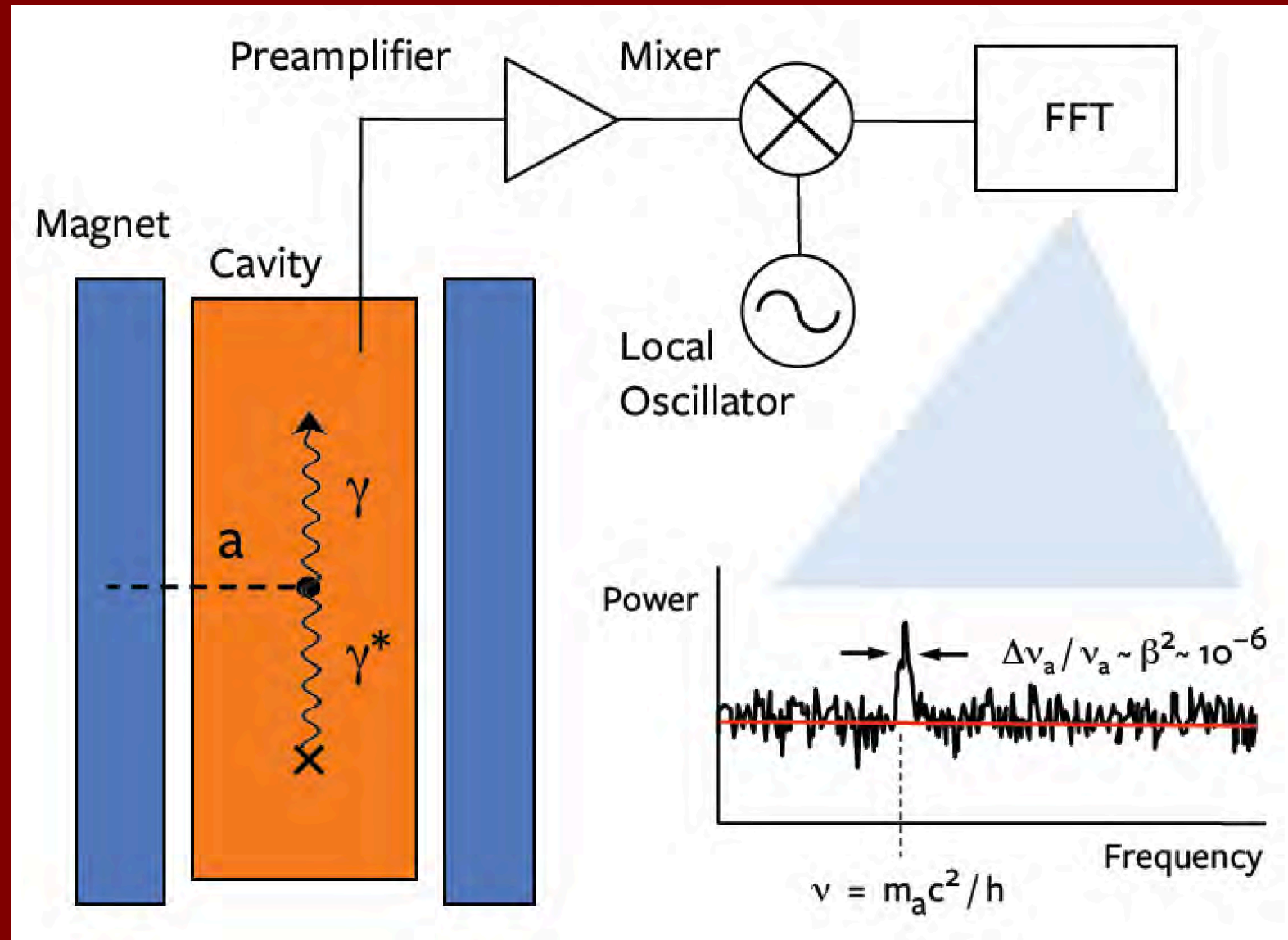


# Helioscope of the future: IAXO



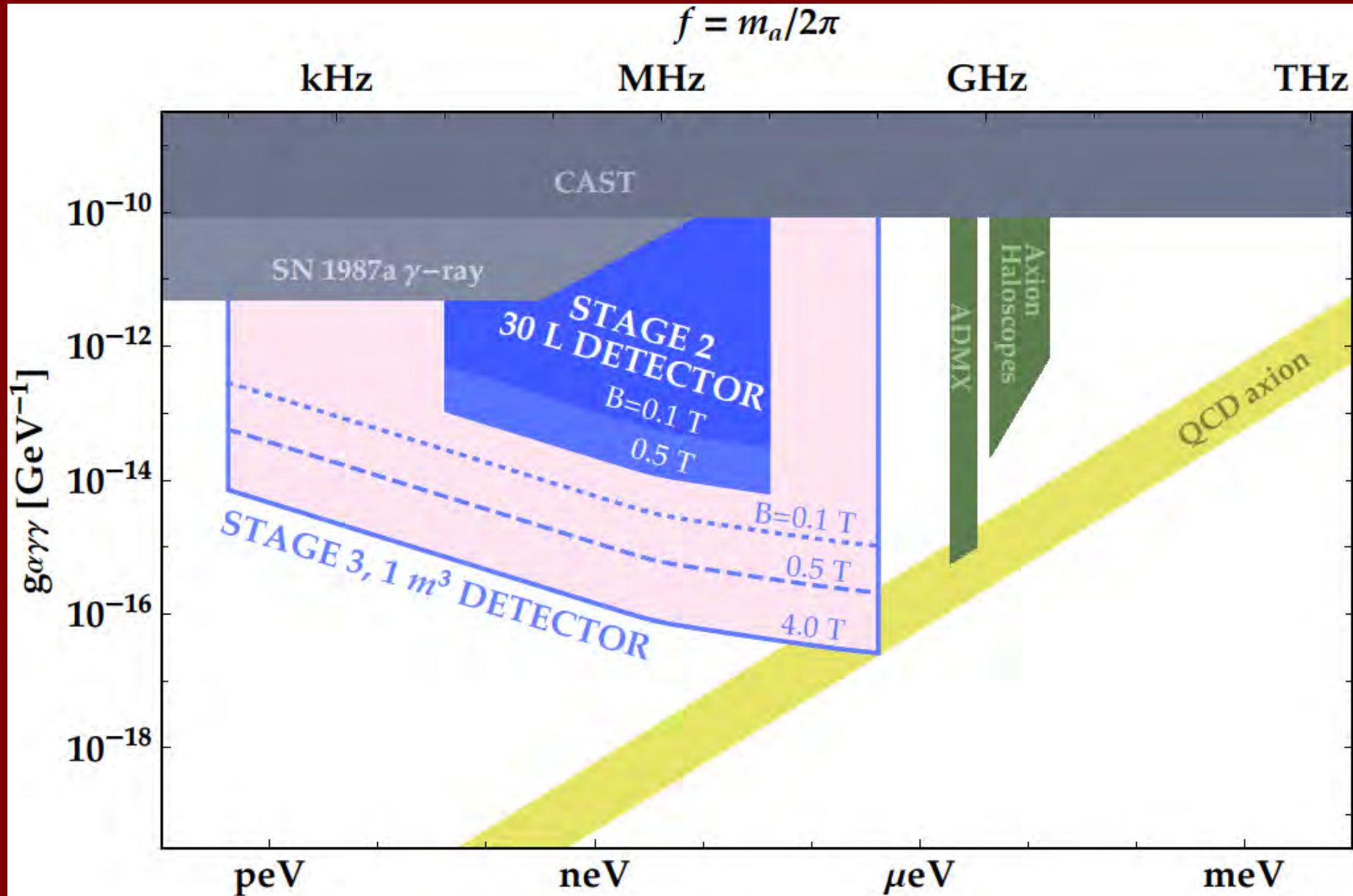


# The principle of the microwave-cavity haloscopes: ADMX, HAYSTACK, CAPP, ORGAN





# New Haloscope Proposals: ABRACADABRA (MIT) and DM Radio



# CASPER-Wind: projected sensitivity

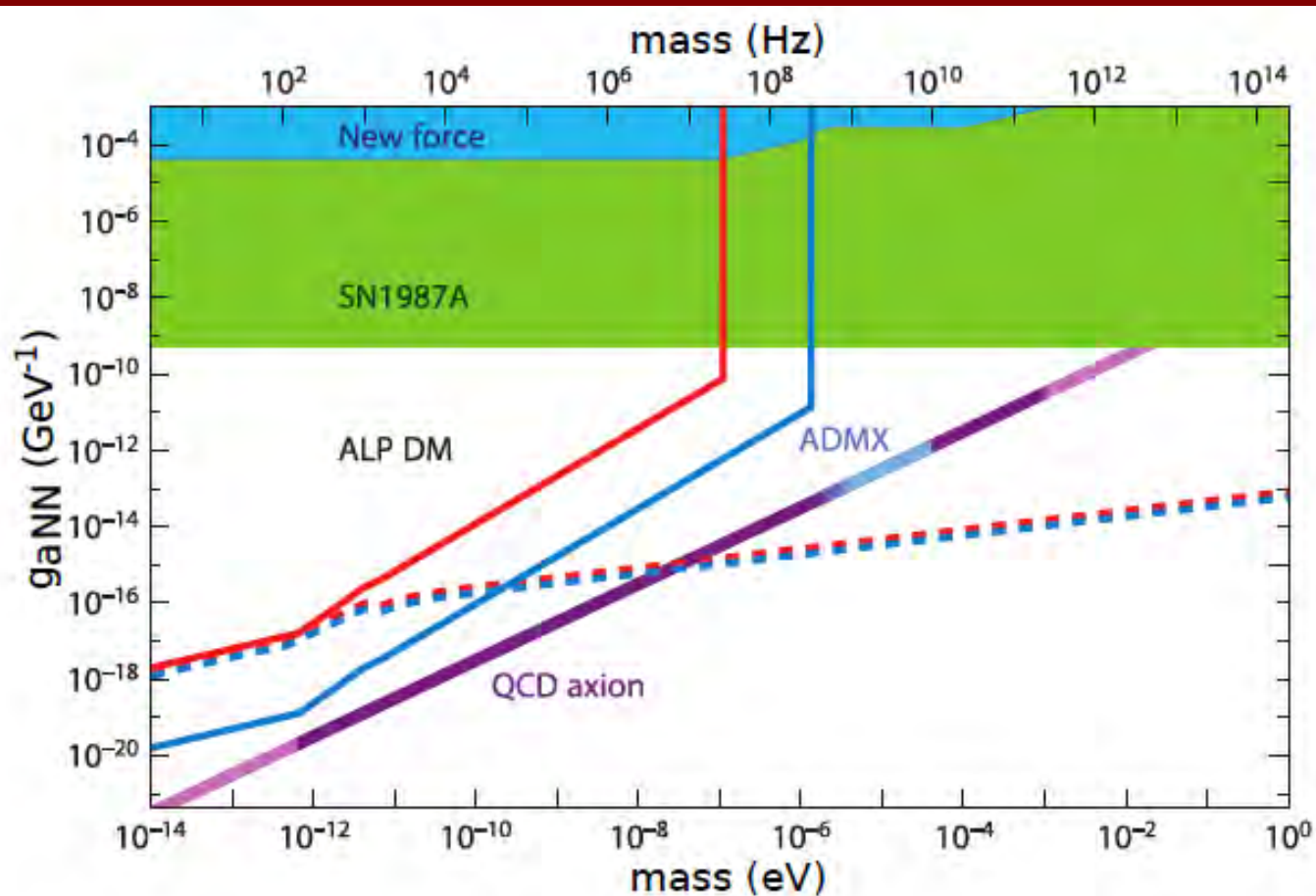


Figure 1: ALP-nucleon coupling parameter space: coupling strength  $g_{aNN}$  versus ALP mass  $m_a$ . The purple line represents the mass-coupling parameter space corresponding to the QCD axion proposed to solve the strong CP problem [25]. The darker purple region of the line shows where the QCD axion could be all of the dark matter. The red line is the projected sensitivity of CASPER-Wind using hyperpolarized  $^{129}\text{Xe}$ . The blue line is the sensitivity using hyperpolarized  $^3\text{He}$  during a future upgrade of the experiment. The dashed lines are the limits from magnetization noise for  $^{129}\text{Xe}$  (red) and  $^3\text{He}$  (blue). The ADMX region shows the mass range already excluded (dark blue) or that will be covered (light blue) by ADMX (probing the axion-photon coupling). The green region is excluded by observations on Supernovae SN1987A [27, 28]. The blue region is excluded by searches for new spin-dependent forces. Figure adapted with permission from Ref. [29].

# The GNOME Experiment

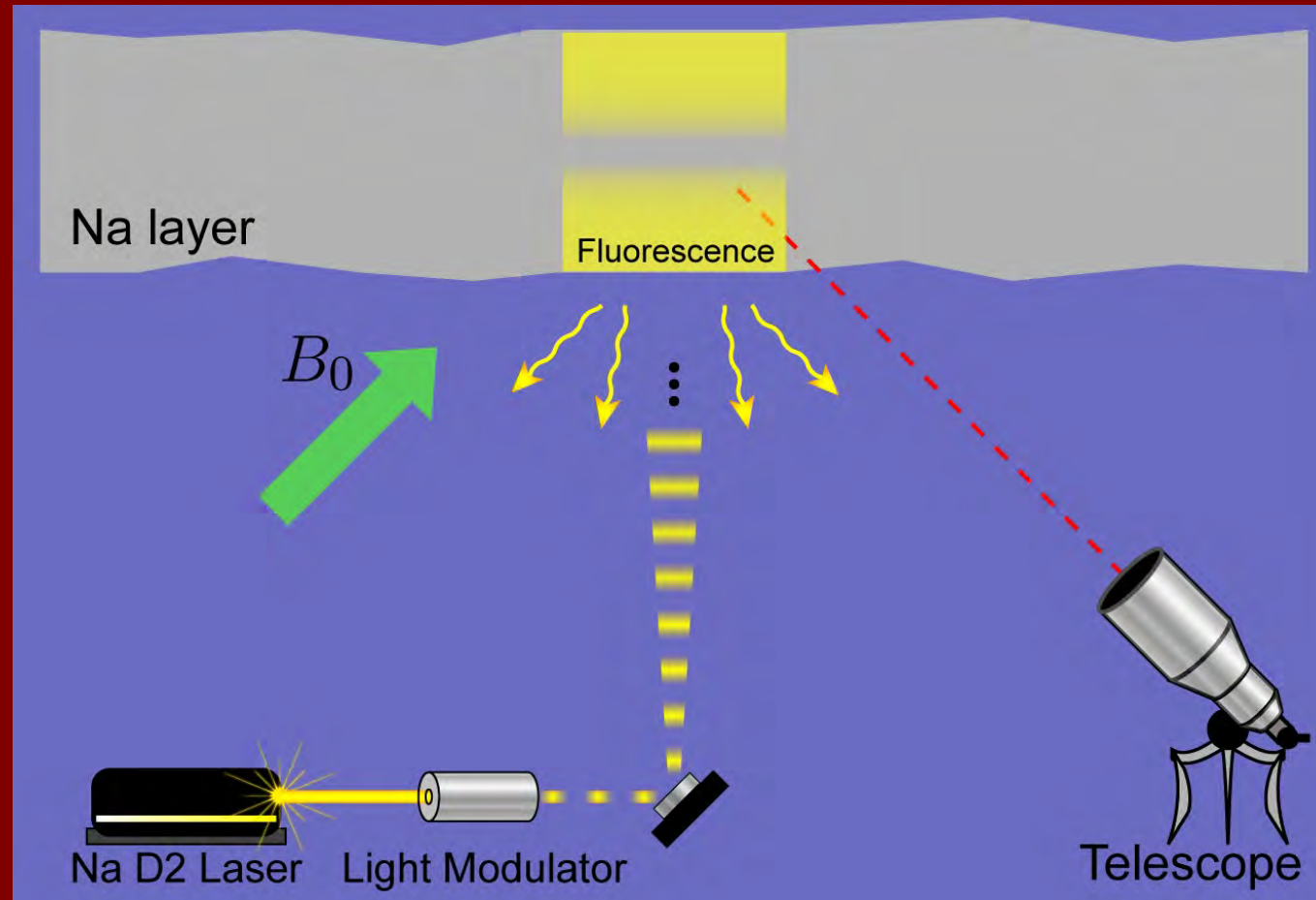
Collaboration website

Current date: 2017/09/28 21:54:36 GPS

Show Map Legend



# Laser Guide-Star magnetometry

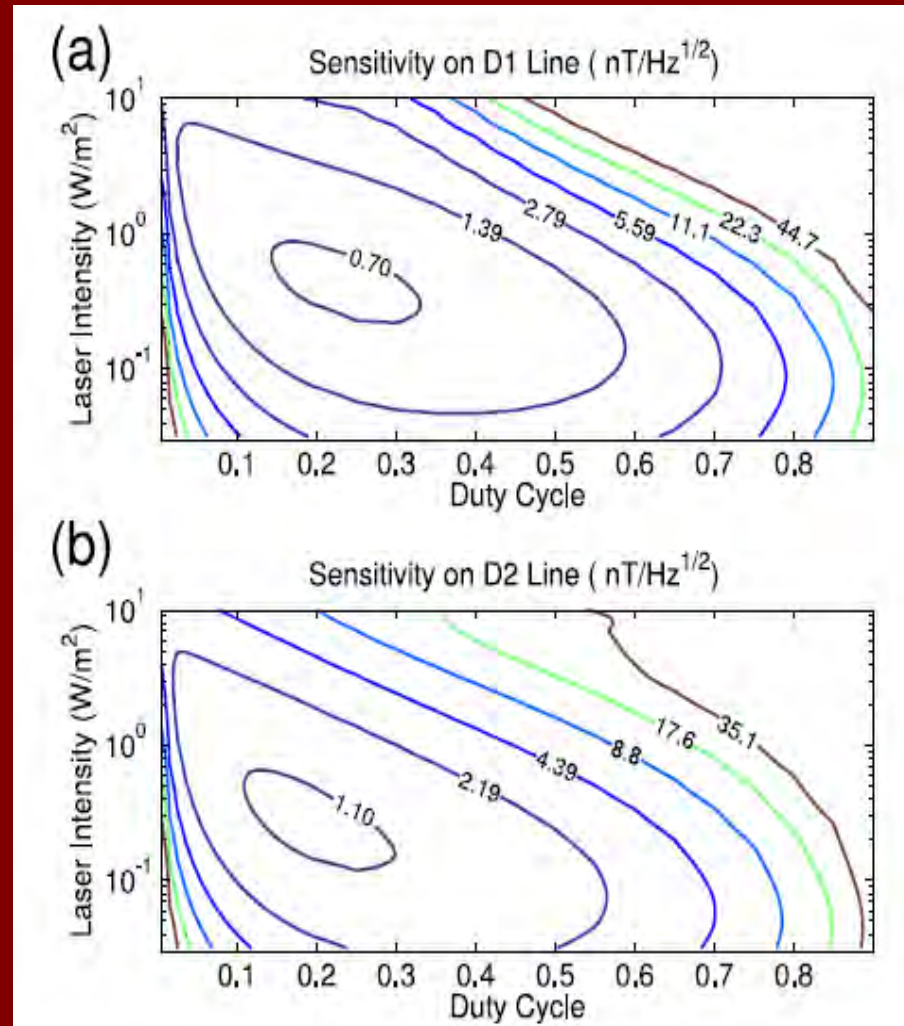
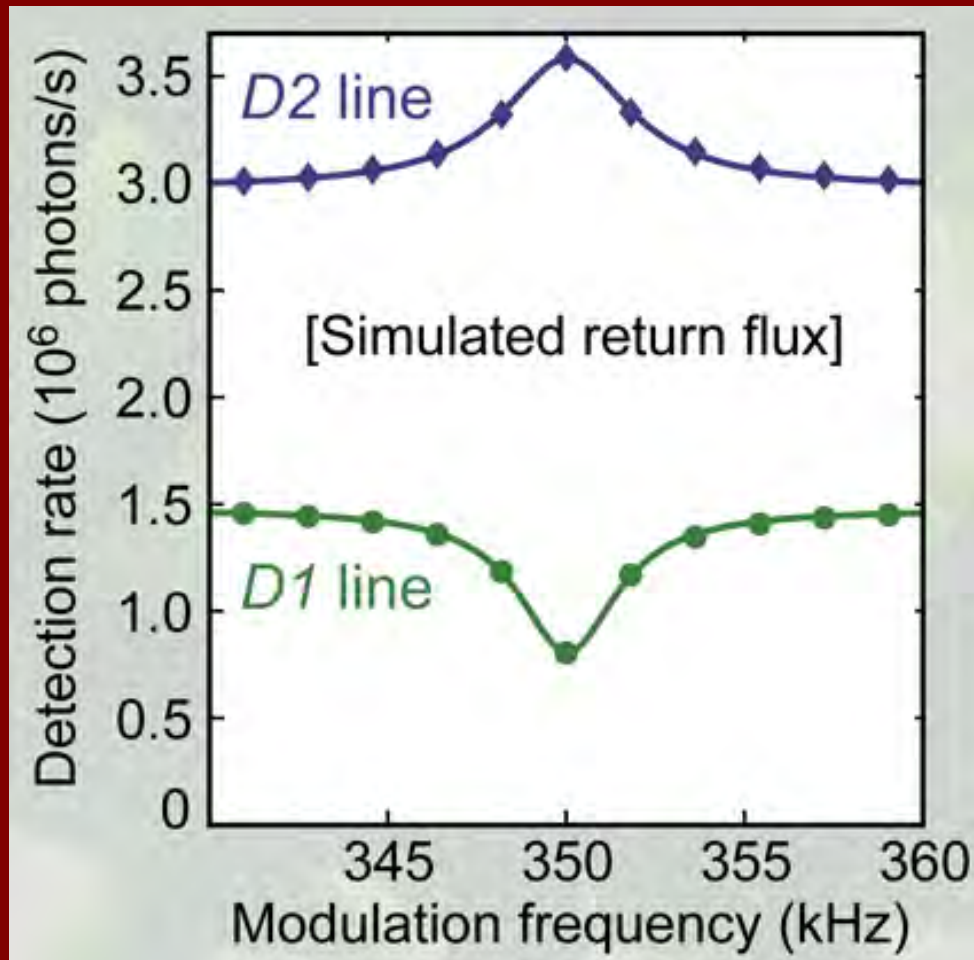


## Magnetometry with mesospheric sodium

James M. Higbie<sup>a,1</sup>, Simon M. Rochester<sup>b</sup>, Brian Patton<sup>b</sup>, Ronald Holzlöhner<sup>c</sup>,  
Domenico Bonaccini Calia<sup>c</sup>, and Dmitry Budker<sup>b,d</sup>

PNAS **108**, 3522 (2011).

# Magneto-optical resonance of mesospheric sodium



# Magnetometer...in the sky!



Dr. D. Bonaccini Calia (ESO)  
Dr. R. Holzlohner (ESO)  
Felipe Pedreros Bustos (Uni-Mainz)

# Magnetometer...in the sky!





Graduate student Georgios Chatzidrosos adjusting an NV-diamond magnetometer



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DMITRY BUDKER &  
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# There is more to do with ZULF NMR!



## Experimental Benchmarking of Quantum Control in Zero-Field Nuclear Magnetic Resonance

Min Jiang, Teng Wu, John W. Blanchard, Guanru Feng, Xinhua Peng, Dmitry Budker

*(Submitted on 21 Aug 2017)*

Zero-field nuclear magnetic resonance (NMR) provides complementary analysis modalities to those of high-field NMR and allows for ultra-high-resolution spectroscopy and measurement of untruncated spin-spin interactions. Unlike for the high-field case, however, universal quantum control -- the ability to perform arbitrary unitary operations -- has not been experimentally demonstrated in zero-field NMR. This is because the Larmor frequency for all spins is identically zero at zero field, making it challenging to individually address different spin species. We realize a composite-pulse technique for arbitrary independent rotations of  $^1\text{H}$  and  $^{13}\text{C}$  spins in a two-spin system. Quantum-information-inspired randomized benchmarking and state tomography are used to evaluate the quality of the control. We experimentally demonstrate single-spin control for  $^{13}\text{C}$  with an average gate fidelity of  $0.9960(2)$  and two-spin control via a controlled-not (CNOT) gate with an estimated fidelity of  $0.99$ . The combination of arbitrary single-spin gates and a CNOT gate is sufficient for universal quantum control of the nuclear spin system. The realization of complete spin control in zero-field NMR is an essential step towards applications to quantum simulation, entangled-state-assisted quantum metrology, and zero-field NMR spectroscopy.

Comments: 19 pages, 3 figures

Subjects: **Quantum Physics (quant-ph)**; Atomic Physics (physics.atom-ph); Chemical Physics (physics.chem-ph)

Cite as: **arXiv:1708.06324 [quant-ph]**