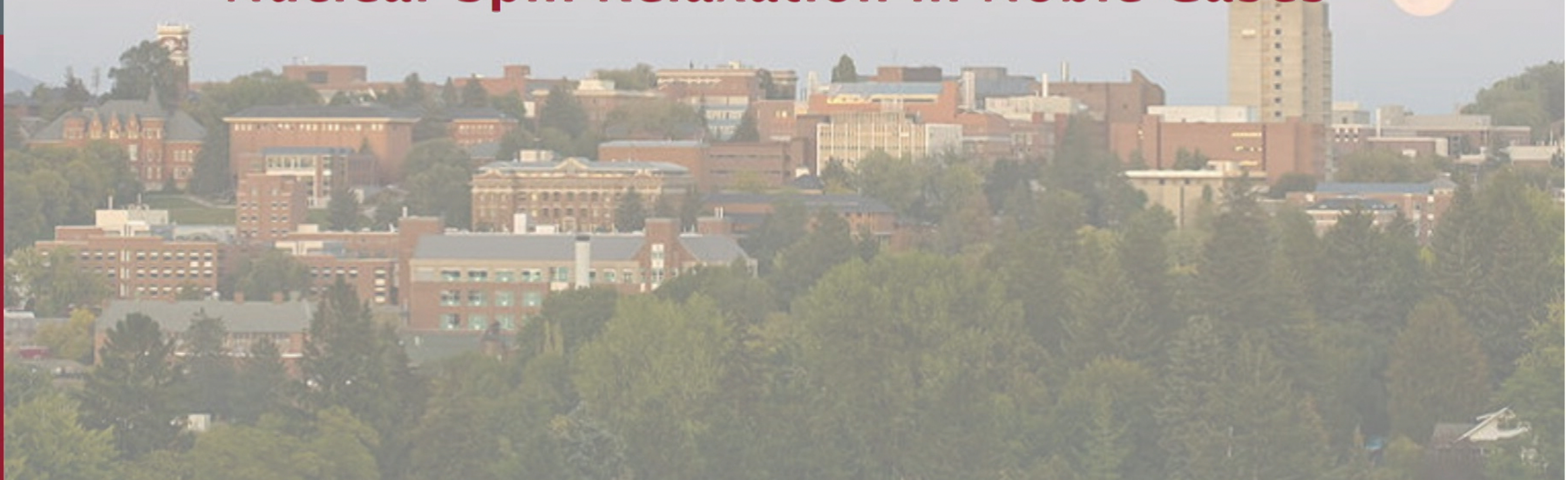
Title: Nuclear Spin Relaxation in Noble Gases

Date: Aug 25, 2017 09:30 AM

URL: http://pirsa.org/17080036

Abstract:

# Nuclear Spin Relaxation in Noble Gases




Brian Saam

Washington State University

Pullman, Washington, USA


*Experimental Techniques in Table-top Fundamental Physics, Perimeter Institute, 25 August 2017*

# Spin Relaxation

- Unperturbed Zeeman Hamiltonian: $\widehat{H}_0 = \mu \cdot \mathbf{B_0}$
- Longitudinal relaxation; time $T_1$
- Transverse ("spin-spin") relaxation; time $T_2$
- Decoherence (not necessarily irreversible); time $T_2^*$
- Hierarchy of times: $T_2^* \leq T_2 \leq T_1$

# *Nuclear* Spin Relaxation

➢ Weak magnetic moments $\mu_N \approx 10^{-3}\mu_B$

# *Gas-Phase* Nuclear Spin Relaxation

- Weak magnetic moments $\mu_N \approx 10^{-3} \mu_B$
- Weak collisional interactions

## *Noble* Gas Nuclear Spin Relaxation

- Weak magnetic moments $\mu_N \approx 10^{-3}\mu_B$
- Weak collisional interactions
- Weak intra-atomic interactions (closed shells)

# *Spin-1/2* Noble Gas Nuclear Spin Relaxation

- Weak magnetic moments $\mu_N \approx 10^{-3}\mu_B$
- Weak collisional interactions
- Weak intra-atomic interactions (closed shells)
- Magnetic-field interactions *only*

# *Spin-1/2* Noble Gas Nuclear Spin Relaxation

- Weak magnetic moments $\mu_N \approx 10^{-3}\mu_B$
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- Weak intra-atomic interactions (closed shells)
- Magnetic-field interactions *only*

- Long $T_1$, $T_2$ (in weak collision limit, $T_2 \to T_1$)

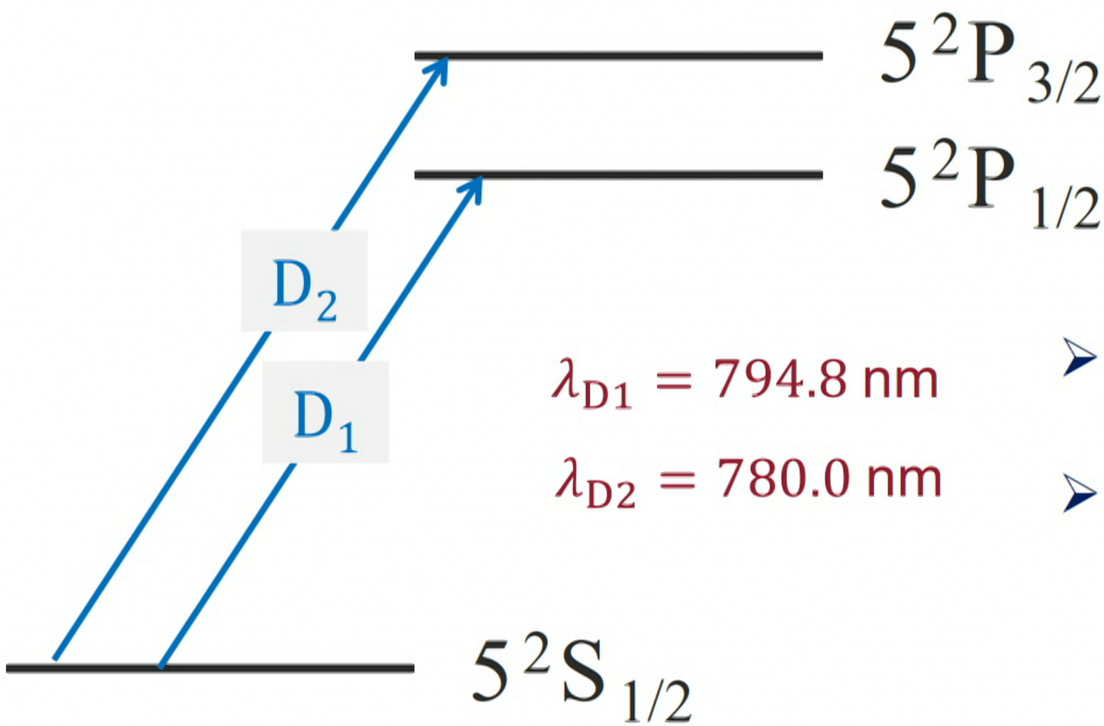
# *Spin-1/2* Noble Gas Nuclear Spin Relaxation

- Weak magnetic moments $\mu_N \approx 10^{-3}\mu_B$
- Weak collisional interactions
- Weak intra-atomic interactions (closed shells)
- Magnetic-field interactions *only*

- Long $T_1$, $T_2$ (in weak collision limit, $T_2 \rightarrow T_1$)
- Allows optical polarization of noble gases, so-called "*hyper*polarization" via spin-exchange or metastability-exchange optical pumping (SEOP or MEOP)

## Rubidium Fine Structure

(ignores Rb nuclear spin)




$\lambda_{D1} = 794.8$ nm

$\lambda_{D2} = 780.0$ nm

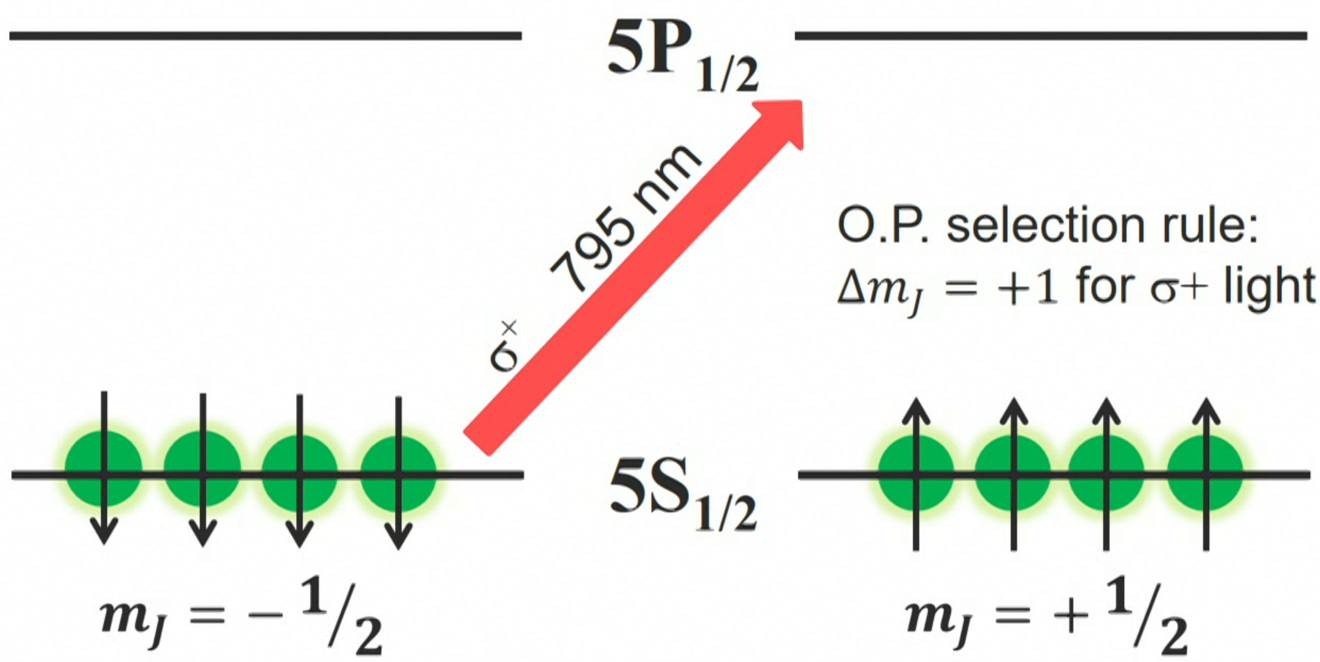
- Single valence electron defines the atomic state
- Selection Rules for Electric Dipole Transitions:

$$\Delta L = \pm 1$$

$$\Delta J = 0, \pm 1$$

# Depopulation Optical Pumping

(still ignoring Rb nuclear spin)



# Collisional Transfer to Noble Gas Nuclei

"Spin Exchange"

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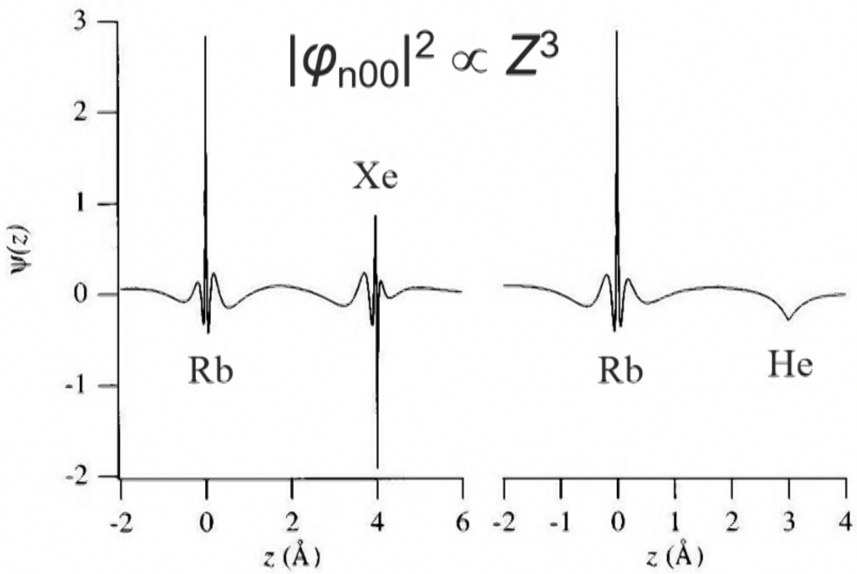
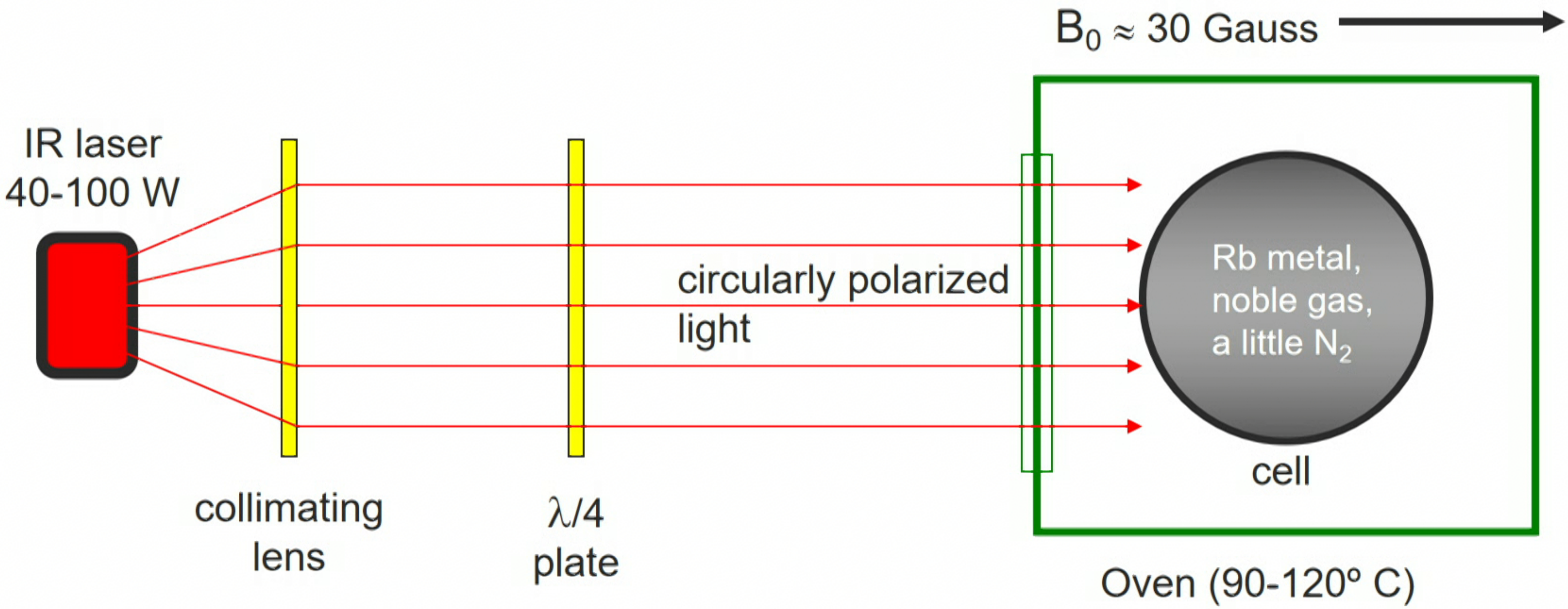


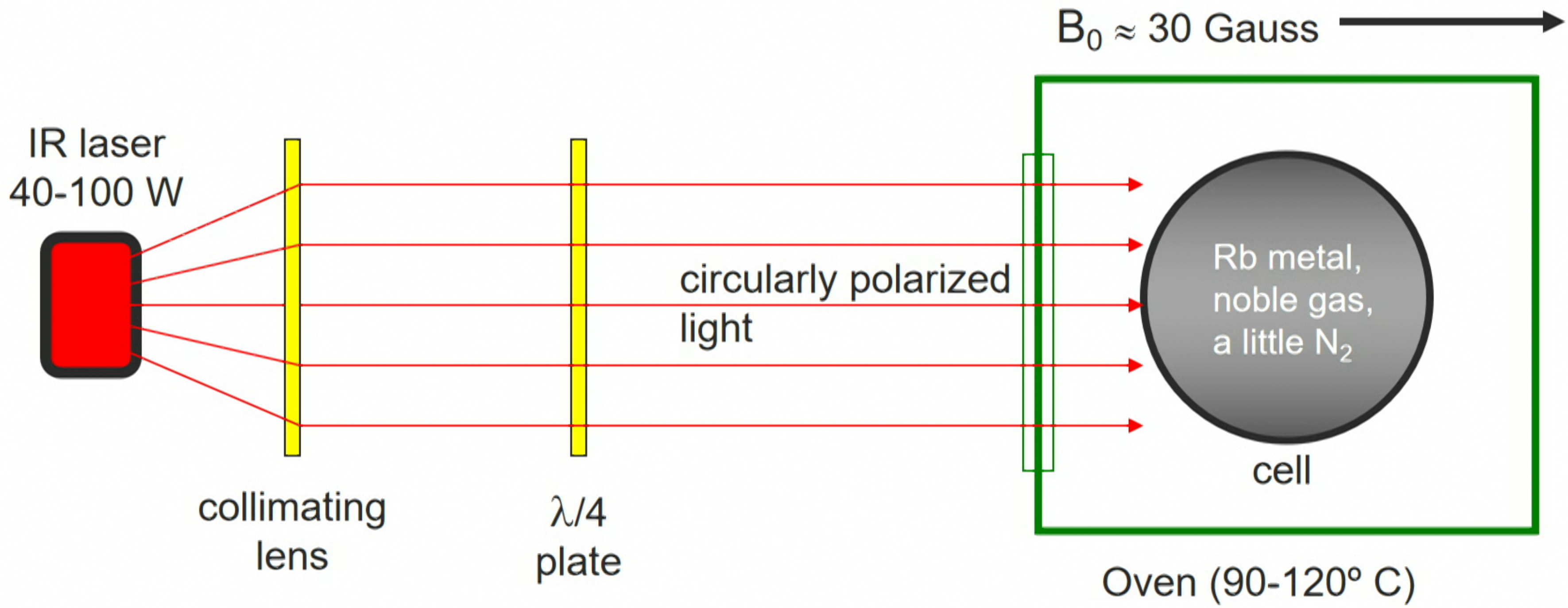

**Rb-Xe interatomic distance**

Walker and Happer, Rev. Mod. Phys. **69**, 629 (1997), Fig. 13.


# SEOP Schematic



# SEOP Schematic



# The Advent of Dynamic Nuclear Polarization (DNP)

## Polarization of Nuclear Spins in Metals*

T. R. CARVER† AND C. P. SLICHTER
*Department of Physics, University of Illinois, Urbana, Illinois*
(Received August 17, 1953)

First experimental demonstration of the Overhauser Effect

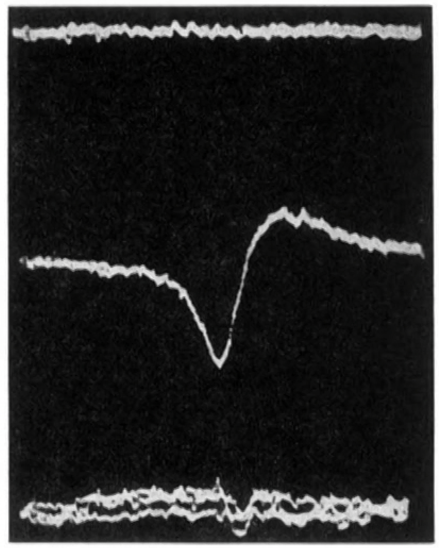


FIG. 1. Oscilloscope pictures of 50-kc/sec nuclear resonance absorption vs static magnetic field. Field excursion 0.2 gauss. Top line: $Li^7$ resonance (lost in noise). Middle line: $Li^7$ resonance enhanced by electron saturation. Bottom line: Proton resonance in glycerin sample.

$^7$Li NMR in lithium metal

- No saturation of conduction electrons
- Electron resonance (only) saturated with strong rf field
- Proton NMR resonance in glycerine sample (for reference)

*Phys. Rev.* **92**, 212 (1953).

# First “Hyperpolarized” Noble Gas

NUCLEAR POLARIZATION IN $He^3$ GAS INDUCED BY OPTICAL PUMPING AND DIPOLAR EXCHANGE*

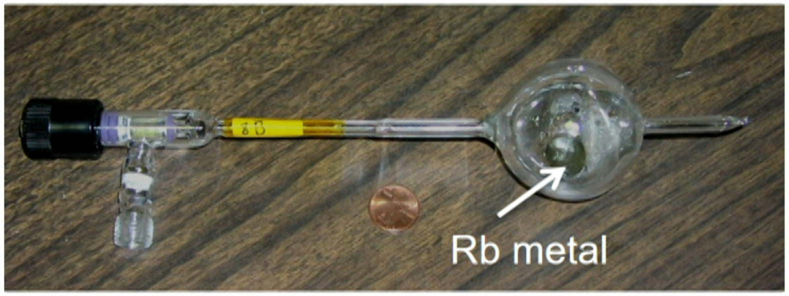
M. A. Bouchiat,† T. R. Carver,‡ and C. M. Varnum
Palmer Physical Laboratory, Princeton University, Princeton, New Jersey
(Received September 26, 1960)

“…the Overhauser effect should work not by the dipole-coupled relaxation of a saturated paramagnetic impurity toward a polarized equilibrium, but by the relaxation of an optically polarized impurity toward a nearly depolarized equilibrium.”

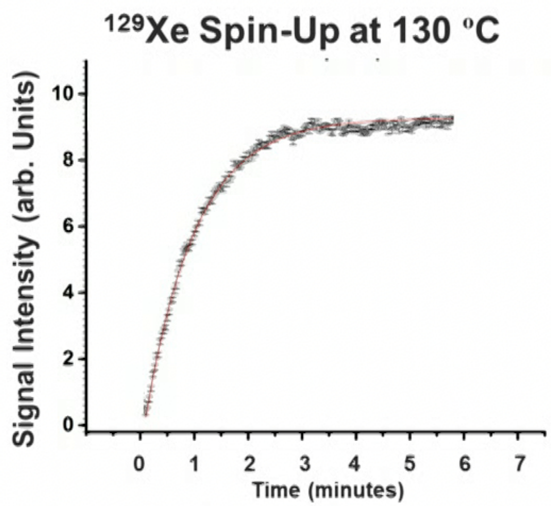
*M.A. Bouchiat, T.R. Carver, and C.M. Varnum, *Phys. Rev. Lett*. **5**, 373 (1960).

# Tools of the Trade

Hyperpolarized $^{129}$Xe "On Tap"




$^{3}$He "Batch-mode" SEOP cell

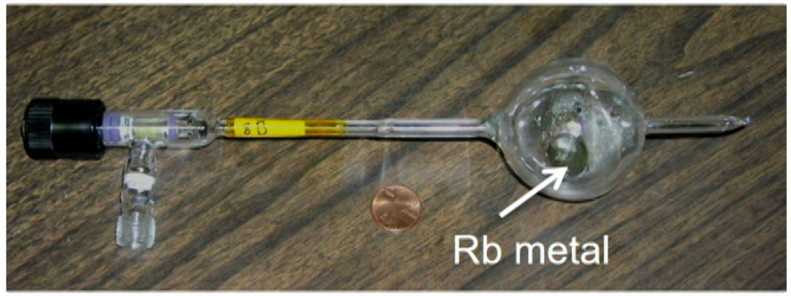



$$P_{\mathrm{Xe}} = \overline{P_{\mathrm{Rb}}}\left(\frac{\gamma_{\mathrm{se}}}{\gamma_{\mathrm{se}} + \Gamma}\right)\left[1 - e^{-(\gamma_{\mathrm{se}}+\Gamma)t}\right]$$

Vacuum/Gas Handling System for Cell Fabrication

# Tools of the Trade

Hyperpolarized $^{129}$Xe
"On Tap"

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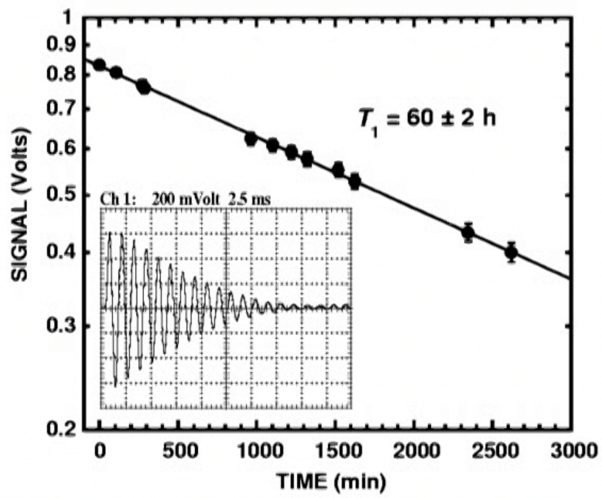


Vacuum/Gas Handling
System for Cell Fabrication

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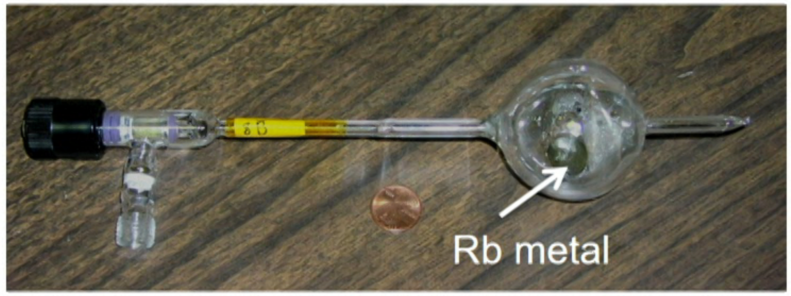


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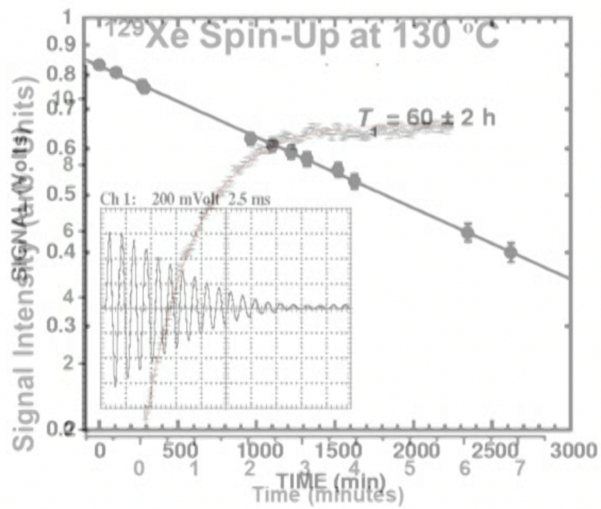


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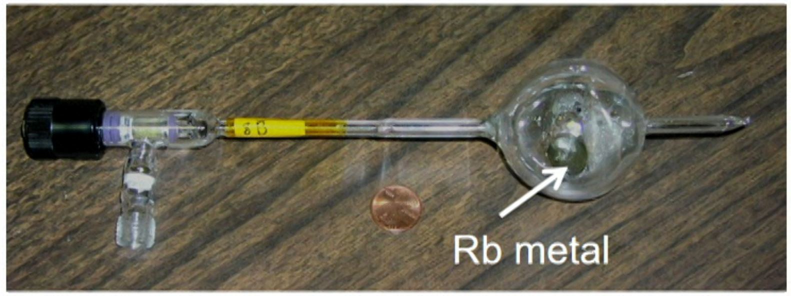


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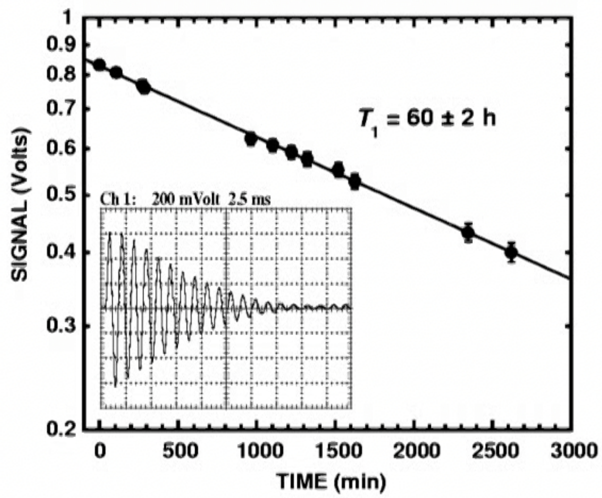
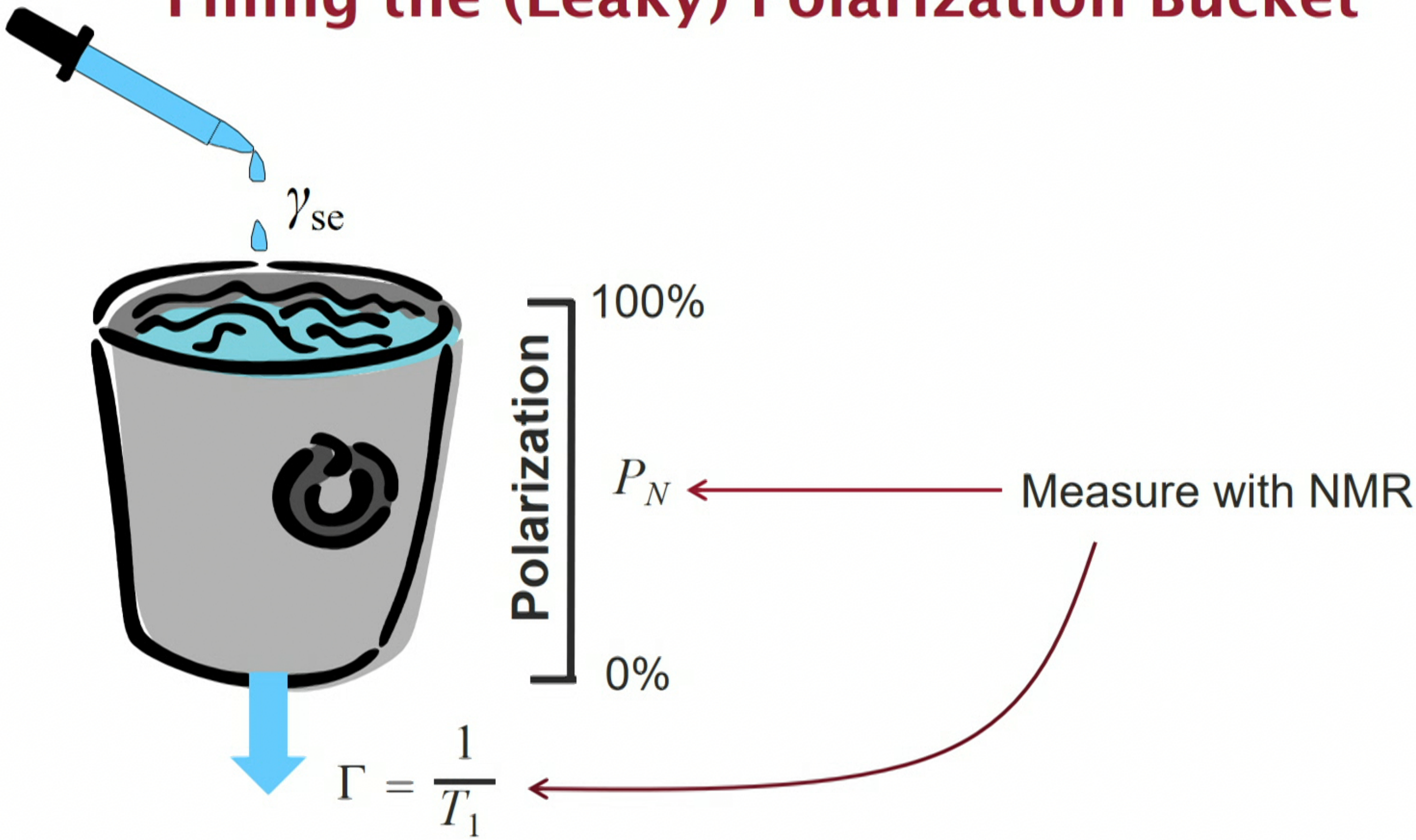
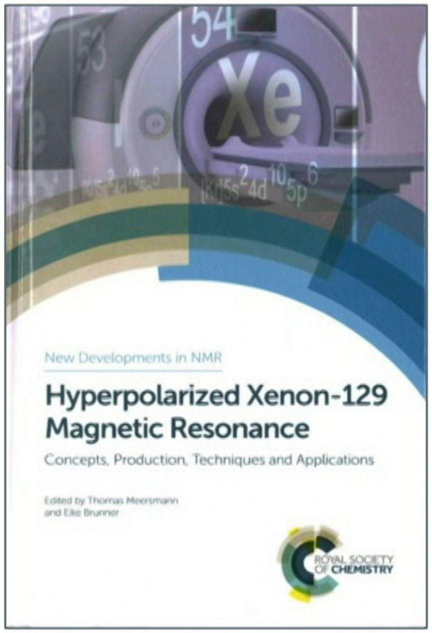



Vacuum/Gas Handling System for Cell Fabrication

# Filling the (Leaky) Polarization Bucket






B. Saam, "$T_1$ Relaxation of $^{129}$Xe and how to keep it long" (Ch. 7) in *Hyperpolarized Xe-129 Magnetic Resonance*, T. Meersman & E. Brunner, eds.

# Reviews

T.R. Gentile, P.J. Nacher, B. Saam, and T.G. Walker, "Optically Polarized $^3$He, Rev. Mod. Phys. (in press, 2017).

## Optically polarized $^3$He

T. R. Gentile
*National Institute of Standards and Technology (NIST), Gaithersburg, Maryland 20899, USA*

P.J. Nacher
*Laboratoire Kastler Brossel, ENS-PSL Research University, CNRS, UPMC-Sorbonne Universités, Collège de France, Paris, France*

B. Saam
*Department of Physics and Astronomy, University of Utah, Salt Lake City, Utah 84112, USA*

T. G. Walker*
*Department of Physics, University of Wisconsin-Madison, Madison, Wisconsin 53706, USA*

(Dated: May 22, 2017)

This article reviews the physics and technology of producing large quantities of highly spin-polarized, or hyperpolarized, $^3$He nuclei using spin-exchange (SEOP) and metastability-exchange (MEOP) optical pumping, and surveys applications of polarized $^3$He. Several recent developments are emphasized for each method. For SEOP, the use of spectrally narrowed lasers and Rb/K mixtures has substantially increased the achievable polarization and polarizing rate. MEOP in high magnetic fields has likewise significantly increased the pressure at which this method can be performed, and has led to the observation of a light-induced relaxation mechanism. In both methods the increased capabilities have led to more extensive study and modeling of the basic underlying physics. New unexplained dependences of relaxation on temperature and magnetic field have been discovered in SEOP cells. Applications of both methods are also reviewed, including targets for charged particle and photon beams, neutron spin filters, magnetic resonance imaging, and precision measurements.

### CONTENTS

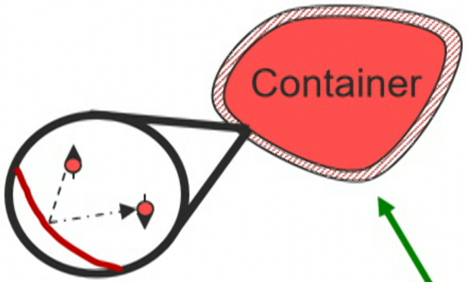


* tgwalker@wisc.edu

# Sources of Longitudinal Relaxation

$$\Gamma = \Gamma_{\text{wall}} + \Gamma_{\text{intrinsic}} + \Gamma_{\text{gradient}} + \Gamma_{\text{oxygen}}$$

# Sources of Longitudinal Relaxation

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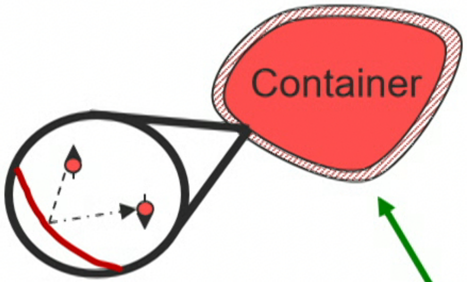


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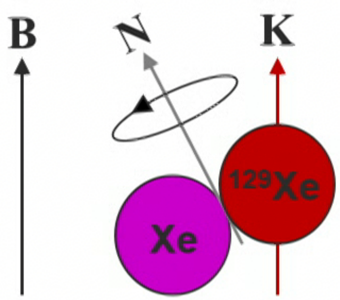


**spin-rotation:**

$$C_K(R)\mathbf{N}\cdot\mathbf{K}$$

**Occurs during binary collision AND during the lifetime of a molecule.**

**For pure Xe:** $\Gamma_{XeXe} \approx 0.2\,\mathrm{hr}^{-1}\mathrm{amgt}^{-1}$

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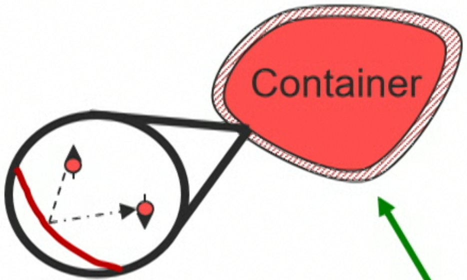


$$\Gamma_{\text{gradient}} = D \frac{|\nabla_\perp B|^2}{B_0^2}$$

**Conventional regime**

G.D. Cates et al., Phys. Rev. A **37**, 2877(1988).

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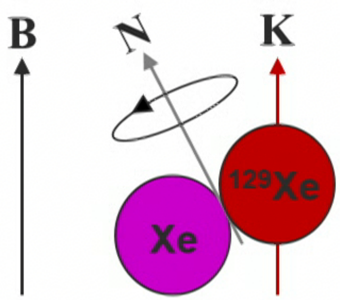


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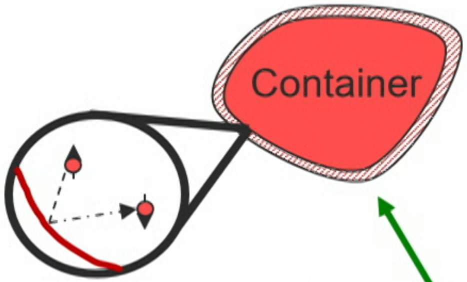


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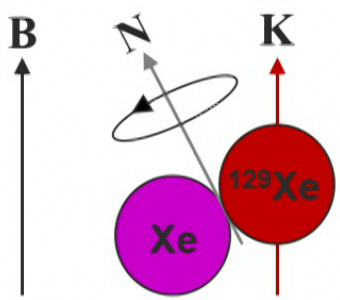


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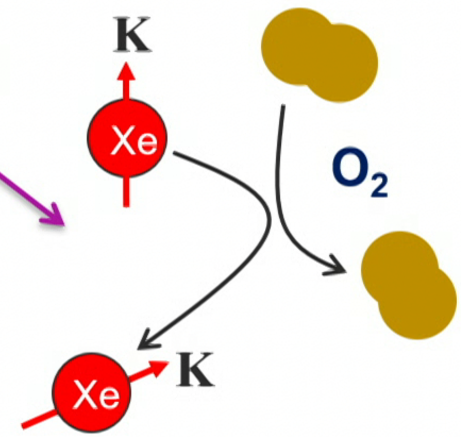
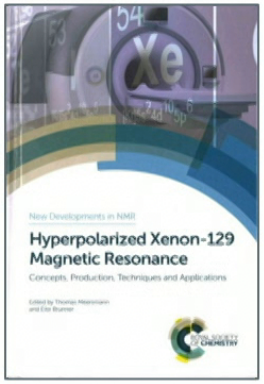



$$\Gamma_{\text{oxygen}} = 0.48\,\text{sec}^{-1}\,\text{amgt}^{-1}[O_2]$$

**For pure Xe:** $\Gamma_{\text{XeXe}} \approx 0.2\,\text{hr}^{-1}\text{amgt}^{-1}$

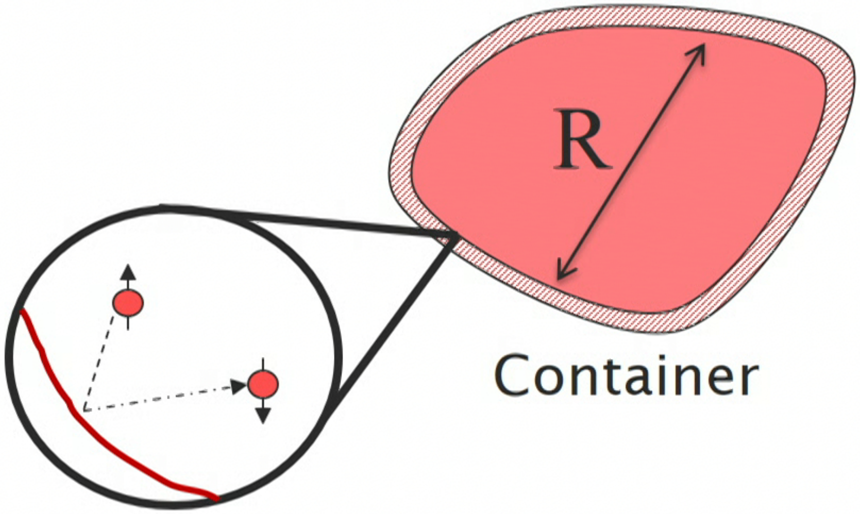
# Noble Gas Relaxation for Spin $K > 1/2$

- Quadrupole moment; relaxation due to E-field gradients (much shorter $T_1$), harder to achieve large polarizations by optical methods
- Quadrupole-broadened line shape can be used as a sensitive probe of chemical environment




T. Meersman & G.E. Pavlovskaya, "Beyond Spin I=1/2; Hyperpolarized $^{131}Xe$ and $^{83}Kr$ Magnetic Resonance" (Ch. 23)

# Wall Relaxation: The Weak-Collision Limit




Relaxation probability
$\alpha$ per wall collision:

$$\alpha << \frac{\lambda}{R}$$

$\lambda$ ← mean free path

$R$ ← Characteristic linear dimension

$$\Gamma_{\text{wall}} = \frac{\alpha \bar{v}}{4}\left(\frac{S}{V}\right)$$ Density independent!!

# Wall Relaxation

"There is perhaps no subject generally pertaining to hyperpolarized noble gases that has been more vexing over the past 50 years than the nature of longitudinal relaxation of nuclei to the walls of the (usually glass) containing vessel or cell. […]The highly deformable electron cloud and the ~0.1 eV van der Waals surface-binding energy of xenon means that wall relaxation times vary widely depending on the size, surface material and/or surface coating, temperature, and applied magnetic field (some who have studied this problem for far too long might also include the phase of the moon)."

-B. Saam, *Hyperpolarized Xenon-129 Magnetic Resonance (Concepts, Production, Techniques and Applications)*

## “We search for islands of reproducibility in a sea of “What the @*&$#???”

-B . Saam, quoted in Ph.D. thesis
of Geoff Shrank

# $^3$He: Aluminosilicate Glasses and Alkali-Metal Coatings

J. Phys. III France 5 (1995) 1287–1295 AUGUST 1995, PAGE 1287

Classification
*Physics Abstracts*
76.60E — 32.80B — 73.60D

**$^3$He Nuclear Spin Relaxation in Cesium Coated Cells at Room Temperature**

B. Chéron([1,2]), H. Gilles([1]), J. Hamel([1]), M. Leduc([3]), O. Moreau([1]) and E. Noël([1])

([1]) Laboratoire de Spectroscopie Atomique, I.S.M.R.A., Bd M[al] Juin, 14050 Caen Cedex, France
([2]) Université de Caen, UFR de Sciences, Esplanade de la paix, 14032 Caen Cedex, France
([3]) Laboratoire Kastler Brossel de l'E.N.S., 24 rue Lhomond, 75231 Paris Cedex 05, France

*(Received 19 January 1995, accepted 16 May 1995)*

## Metastability exchange optical pumping (MEOP) cells

N·H ELSEVIER 29 May 1995 PHYSICS LETTERS A
Physics Letters A 201 (1995) 337–343

**Very long nuclear relaxation times of spin polarized helium 3 in metal coated cells**

Werner Heil [a], Hubert Humblot [a], Ernst Otten [a], Matthias Schafer [a], Reinhard Sarkau [a], Michèle Leduc [b]

[a] *Institut für Physik der Universität Mainz, D-55099 Mainz, Germany*
[b] *Laboratoire Kastler Brossel, ENS, 24 Rue Lhomond, 75231 Paris Cedex 05, France*

Received 17 February 1995; accepted for publication 23 February 1995
Communicated by B. Fricke

$T_1 > 100$ hours
(dominated by wall relaxation)

# Persistence and Statistics

$^3$He Cells: Aluminosilicate Neutron-Spin-Filter Cells at NIST

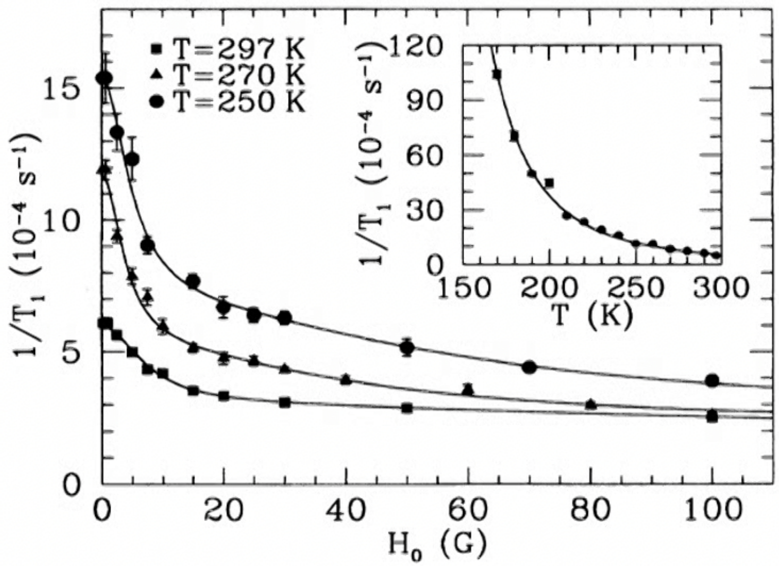
| Cell | $D$ | $d$ | $l$ | $V$ | $p$ | $T_1$ | $P_{He}(795)$ | $P_{He}(770)$ |
|---|---|---|---|---|---|---|---|---|
| Chardonnay | 1.6 | 8.4 | 7.3 | 400 | 1.07 | 390 | 0.77 | 0.67 |
| Orvieto | 2.2 | 8.7 | 7.2 | 430 | 1.1 | 80 | 0.70 | 0.63 |
| Zinfandel | 1.0 | 11.6 | 8.9 | 940 | 1.86 | 330 | 0.78 | |
| Chianti | 2.0 | 11.8 | 8.1 | 875 | 1.82 | 290 | 0.75 | |
| Barbera | 4.4 | 11.8 | 7.5 | 820 | 1.52 | 310 | 0.76 | |
| Syrah | 6.2 | 10.2 | 9.7 | 790 | 1.43 | 450 | 0.76 | 0.73 |
| Riesling | 4.8 | 9.8 | 9.0 | 680 | 3.5 | 170 | 0.77 | 0.68 |
| Cabernet | 46 | 11.8 | 7.6 | 830 | 1.94 | 270 | 0.58 | 0.71 |
| Gigantor | 96 | 9.1 | 9.4 | 611 | 0.85 | 800 | 0.56 | 0.73 |
| Quasimoto | 155 | 6.7 | 5.8 | 205 | 1.12 | 550 | 0.48 | 0.76 |
| Sonora | ∞ | 4.5 | 4.3 | 68 | 0.81 | 500 | | 0.77 |
| Wilma | 0 | 4.9 | 4.9 | 91 | 0.9 | 830 | 0.76 | |
| Chekhov | 0 | 8.1 | 6.0 | 309 | 1.27 | 340 | 0.79 | |
| Bullwinkle | 0 | 6.9 | 7.1 | 268 | 1.22 | 550 | 0.72 | |
| Dino | 0 | 10.6 | 5.1 | 450 | 0.85 | 530 | 0.75 | |
| Kirk | 0 | 10.5 | 7.2 | 620 | 0.80 | 740 | 0.77 | |

$T_1$ in **hours** !!

$T_1$ = 4.9 weeks, limited by $^3$He-$^3$He dipolar mechanism.

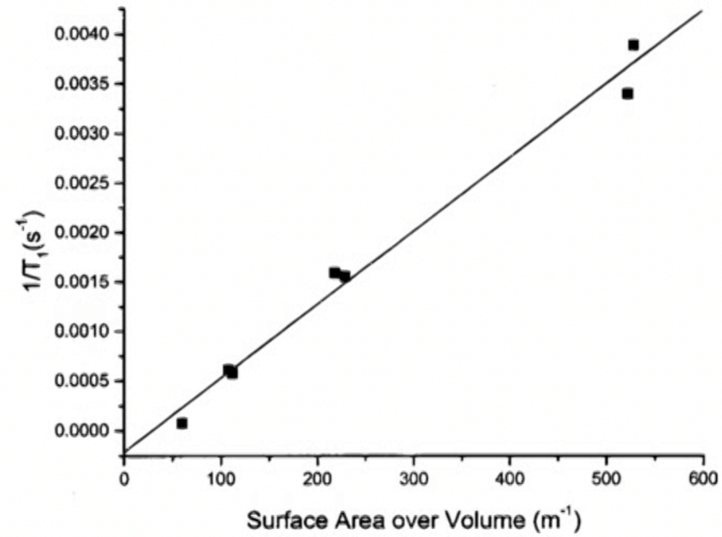
W.C. Chen, T.R. Gentile, T.G. Walker, and E. Babcock, Phys. Rev. A **75**, 013416 (2007).

# $^{129}$Xe: Polymer Coatings to Inhibit Wall Relaxation




B. Driehuys et al., Phys. Rev. Lett. **74**, 4943 (1995).

- 1 cm$^3$ Rb-Xe cells coated with SurfaSil
- Double resonance expt established that protons relax Xe in the coating
- Need to have at least 20 G applied field with coated cells.

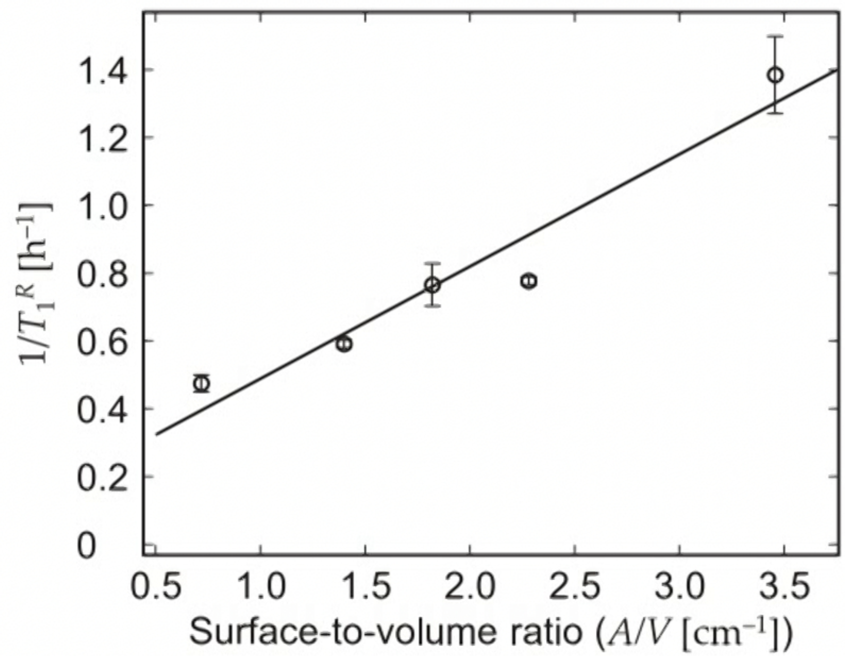



S.R. Breeze, et al., J. Appl. Phys. **74**, 4943 (1995).

- Tried several coatings, incl. Dotriacontane
- $T_1$ = 2-3 hr in applied field of 4.7 T.
- Need to have at least 20 G applied field with coated cells.

# Tedlar™ Bags for Magnetic Resonance Imaging

Relaxation of hyperpolarized $^{129}Xe$ in a deflating polymer bag (Tedlar™)




H.E. Möller *et al.*, J. Magn. Reson. **212** 109-115 (2011).

# State of the Art for $^{129}$Xe Wall Times

Contents lists available at ScienceDirect

Journal of Magnetic Resonance

journal homepage: www.elsevier.com/locate/jmr

Systematic $T_1$ improvement for hyperpolarized $^{129}$xenon

Maricel Repetto [a], Earl Babcock [b], Peter Blümler [a,*], Werner Heil [a], Sergei Karpuk [a], Kathlynne Tullney [a]

[a] Institute of Physics, Johannes Gutenberg University, Staudingerweg 7, 55128 Mainz, Germany

[b] Jülich Centre for Neutron Science, Forschungszentrum Jülich GmbH, Outstation at MLZ, Lichtenbergstrasse 1, 85747 Garching, Germany

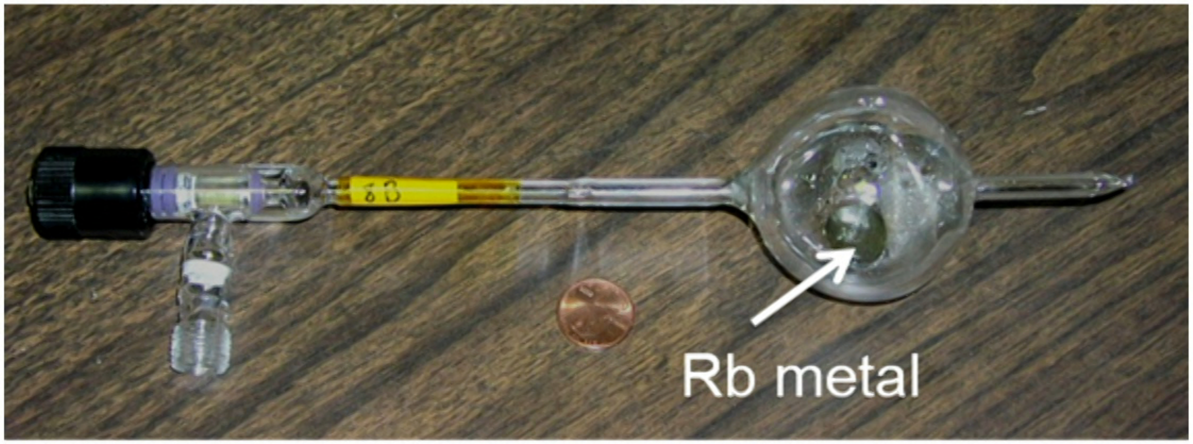
J. Magn. Reson. 252, 163-169 (2015).

$T_1$ up to 18 hours (!)

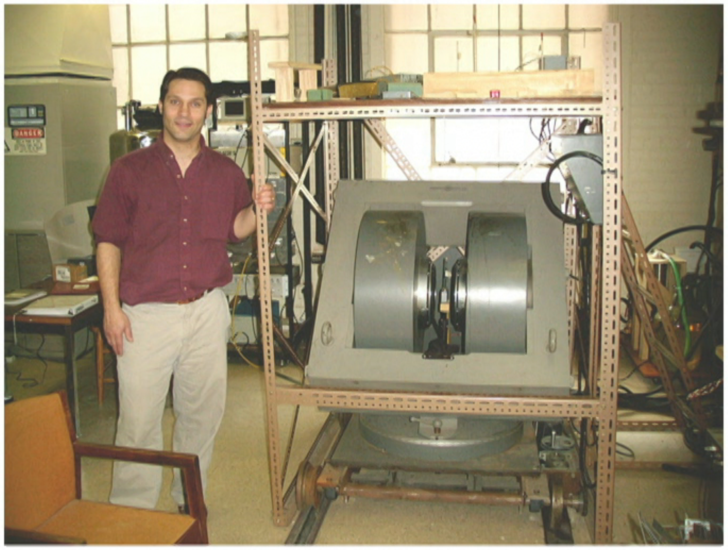
- 10 cm diam spherical cells; GE-180 (aluminosilicate) glass;
- No alkali-metal in cell; **no coating**; valved and re-usable cell
- Low homogeneous (few-gauss) applied field
- Use of additional $N_2$ to shut down $Xe_2$ persistent dimers
- Meticulous wall-cleaning procedure; cell degrades but can be recleaned

# Two Adventures in the Study of Spin Relaxation

## I. Ferromagnetic glass walls



# A funny thing happened on the way to the electromagnet…



$^{3}$He Polarimetry: Compare thermal proton NMR signal with HP $^{3}$He signal in a large magnetic field (1 Tesla).

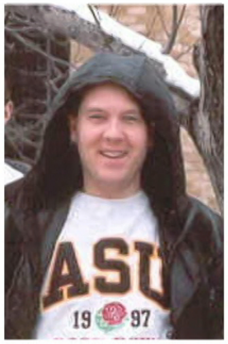
$$P_p = \frac{\mu B}{k_B T}$$

$$\frac{S_{He}}{S_p} = \frac{n_{He}}{n_p}\left(\frac{\gamma_{He}}{\gamma_p}\right)^2 \frac{P_{He}}{P_p}$$

# “Hey, Boss! How many of these things do you want me to ruin?”

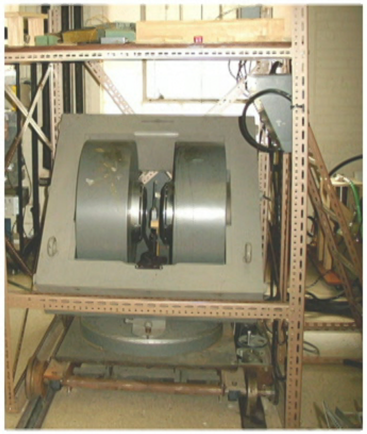
$B_0$ = 30 Gauss


1. Measure $T_1$ = 50 h




Rick Jacob

$B_0$ = 1 Tesla



2. Measure $P_{He}$

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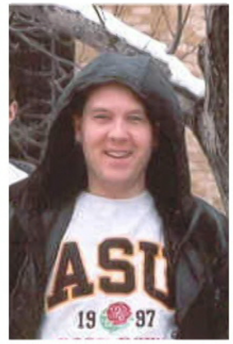
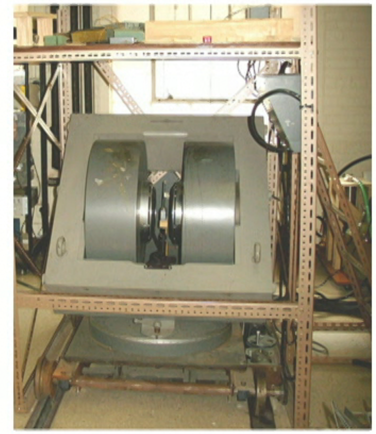



Rick Jacob

$B_0$ = 1 Tesla



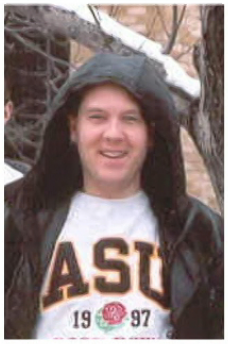
1. Measure $T_1$ = 50 h
2. Measure $P_{He}$
3. Measure $T_1$ = 5 h

# "Hey, Boss! How many of these things do you want me to ruin?"

$B_0$ = 30 Gauss

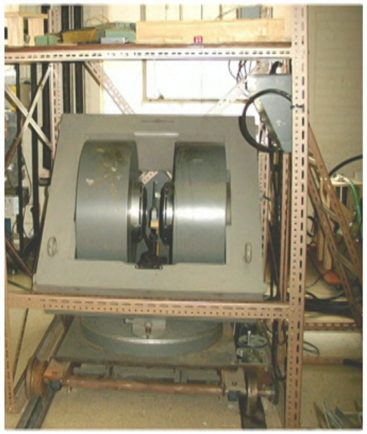

1. Measure $T_1$ = 50 h

3. Measure $T_1$ = 5 h




Rick Jacob

$B_0$ = 1 Tesla



2. Measure $P_{He}$

4. "Degauss"

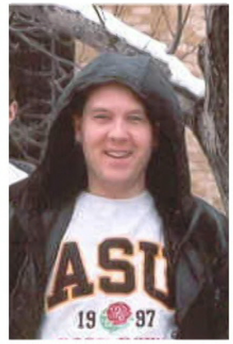
# "Hey, Boss! How many of these things do you want me to ruin?"

$B_0$ = 30 Gauss

1. Measure $T_1$ = 50 h

3. Measure $T_1$ = 5 h

5. Measure $T_1$ = 50 h




Rick Jacob

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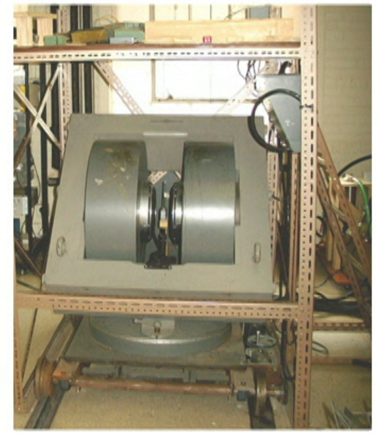


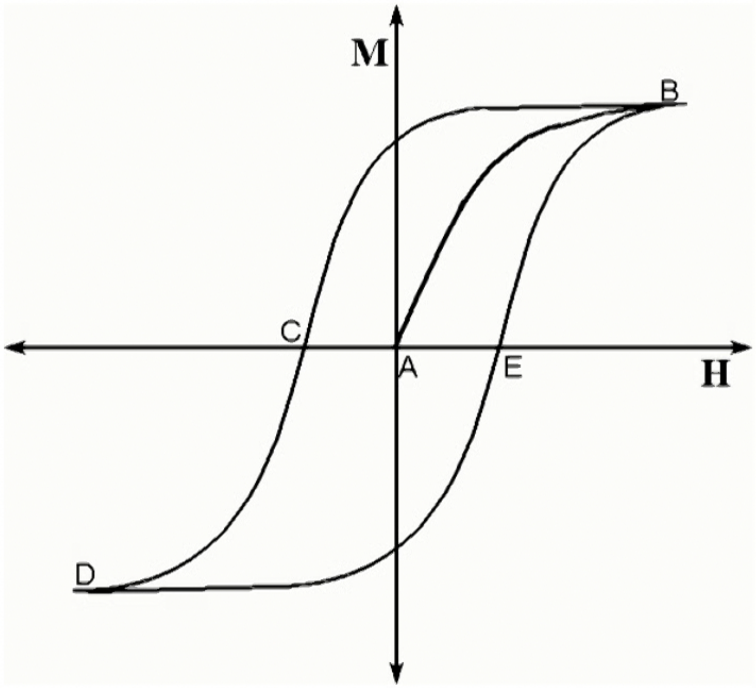
2. Measure $P_{He}$

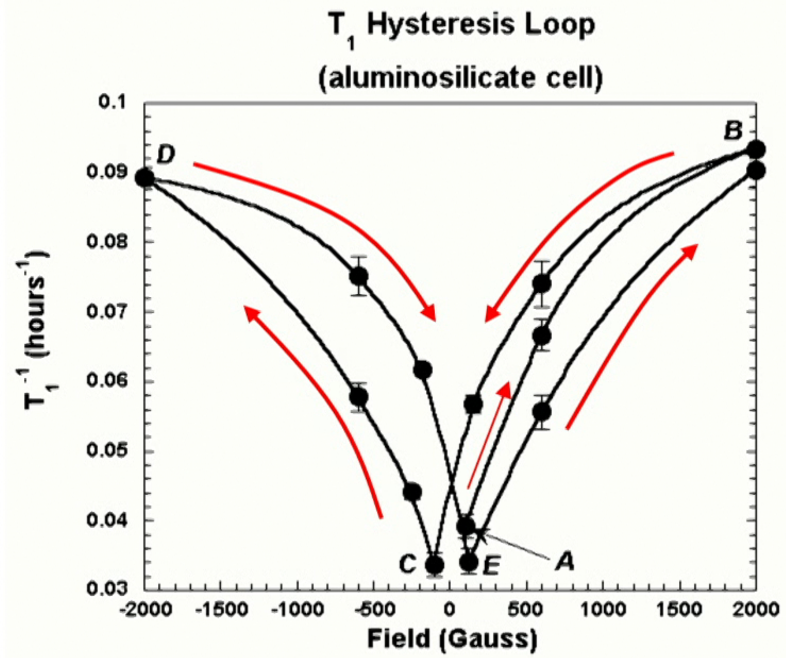
4. "Degauss"

And so on…

➢ Large change in wall relaxation due solely to *previous exposure* to a large magnetic field.

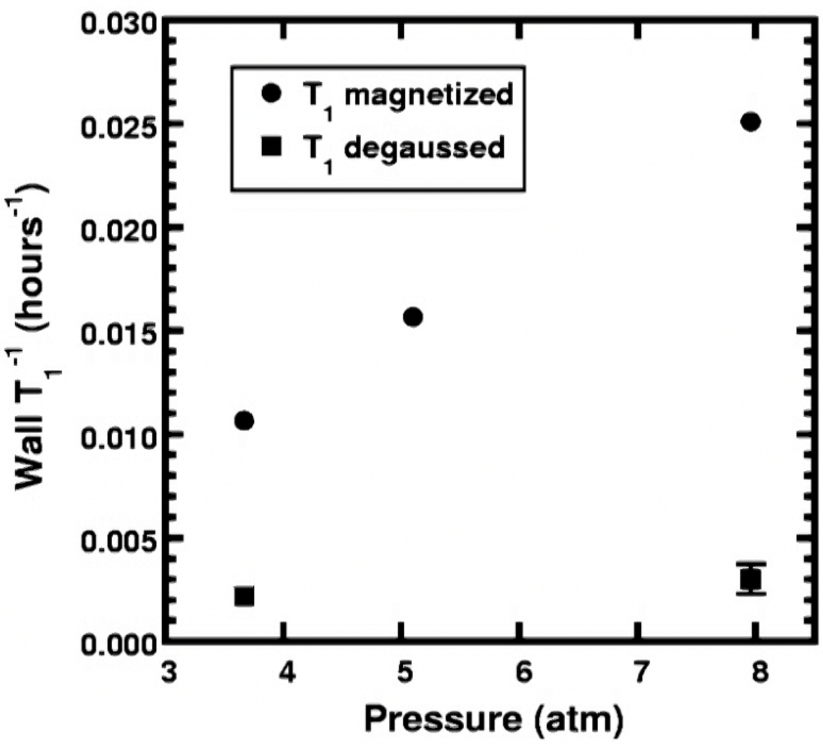
# "$T_1$ Hysteresis"



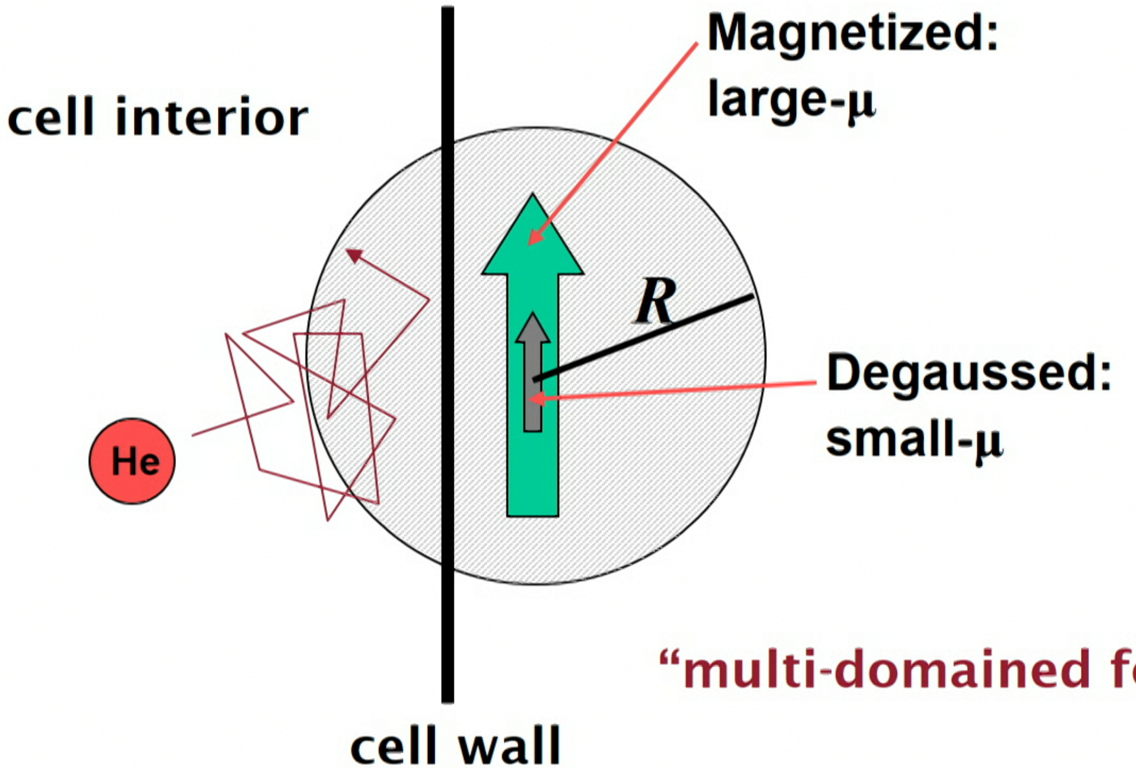

A Origin
B,D Saturation fields
C,E Coercive fields

# Wall Relaxation is Density Dependent for *Magnetized* Cells




$T_1^{-1}$ is linear in pressure for **magnetized** cells only.

# So What's Happening at the Cell Wall?




**Weak collision limit:**

$$\frac{1}{T_1} = M_2\tau$$

$$\frac{1}{T_1} = \frac{N\pi\gamma^2\mu^2}{9RDV}$$

"multi-domained ferromagnetic inclusion"

RE Jacob, SW Morgan, B. Saam, and J.C. Leawoods, Phys. Rev. Lett. 87, 143004 (2001).

# The Mainz Trilogy on $^3$He Wall Relaxation

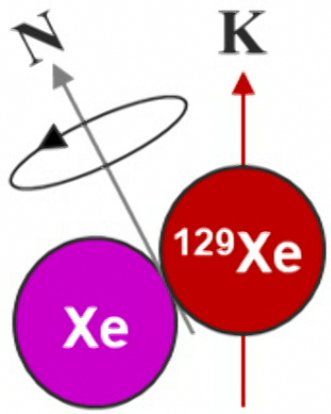
- J. Schmiedeskamp, W. Heil, E.W. Otten, et al., "Paramagnetic relaxation of spin polarized $^3$He at bare glass surfaces: Part III," Eur. Phys. J. D **38** (3), 427–438 (2006).

- A. Deninger, W. Heil, E.W. Otten, *et al.*, "Paramagnetic relaxation of spin polarized $^3$He at coated glass walls: Part II," Eur. Phys. J. D **38** (3), 439–443 (2006).

- J. Schmiedeskamp, H.-J. Elmers, W. Heil, E.W. Otten, *et al.*, "Relaxation of spin polarized $^3$He by magnetized ferromagnetic contaminants: Part III," Eur. Phys. J. D **38** (3), 445-454 (2006).

# Two Adventures in the Study of Spin Relaxation

## II. Intrinsic Relaxation of $^{129}$Xe




A one-atmosphere room-temperature sample of xenon gas has about 1% of its atoms bound in van der Waals dimers; the molecular lifetime is ~ 1 ns.

N. Bernardes and H. Primakoff,
J. Chem. Phys. 30, 691 (1959).

# Early Studies of Intrinsic $^{129}$Xe Relaxation

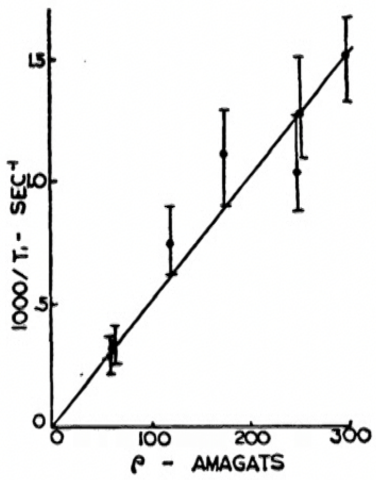



FIG. 3. The $Xe^{129}$ relaxation rate $1/T_1$ in the gas at 25°C as a function of density $\rho$.

$$\Gamma \propto [\mathrm{Xe}]$$

E.R. Hunt and H.Y. Carr,
*Phys. Rev.* **130**, 2302 (1963).

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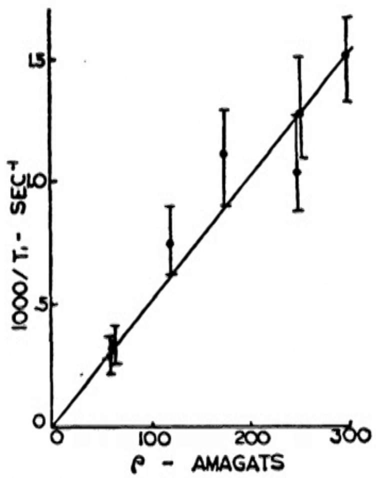


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- *Lowest* density studied is [Xe] = 50 amagats.

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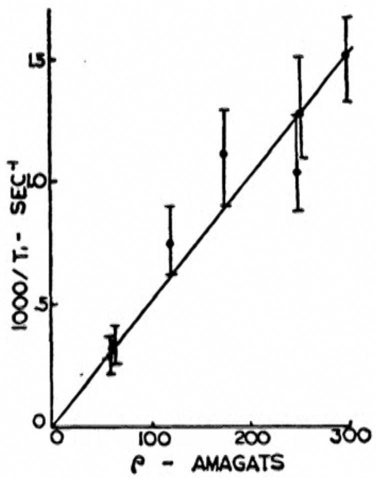


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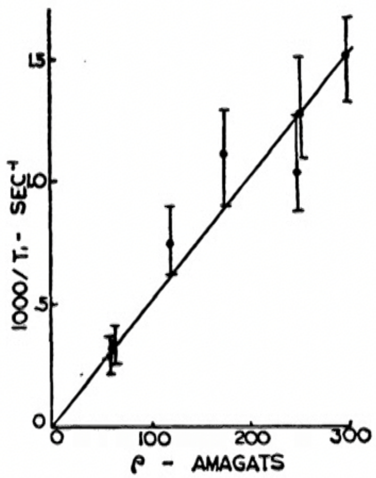


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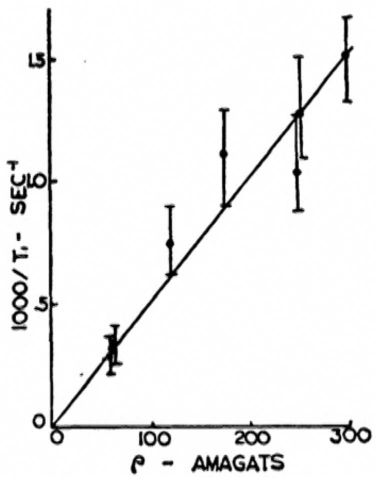
# Early Studies of Intrinsic $^{129}$Xe Relaxation

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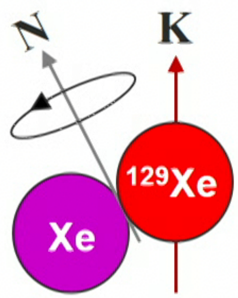
- *Lowest* density studied is [Xe] = 50 amagats.
- Predicts liquid/supercritical $T_1$'s fairly well
- Predicts $T_1 \approx 56$ h for 1 amagat Xe
- We don't need to worry about intrinsic relaxation ever again.

# Transient vs. Persistent Dimers

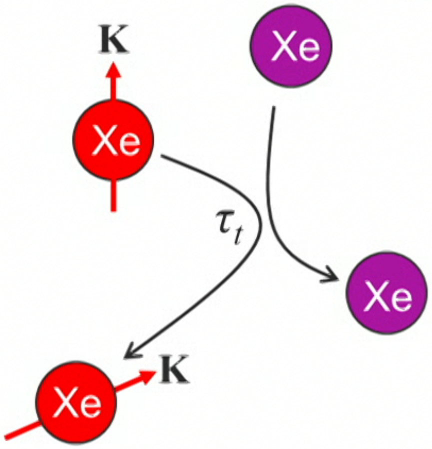
$$\Gamma_{\text{intrinsic}} = \Gamma_t + \Gamma_p$$

**Spin-rotation interaction**:




$$C_K(R)\mathbf{N} \cdot \mathbf{K}$$

# Transient vs. Persistent Dimers

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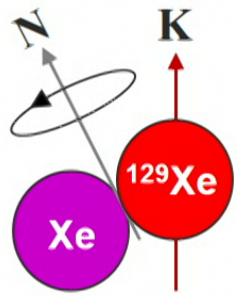


$$C_K(R)\mathbf{N} \cdot \mathbf{K}$$

## TRANSIENT DIMERS ($t$)

- Binary collision time $\tau_t \approx 1$ ps
- $\Gamma_t \propto [\mathrm{Xe}]$
- $\Gamma_t^{-1} \approx 56$ hours·amagat*

* Hunt and Carr (1963); Moudrakovski, et al. (2001)

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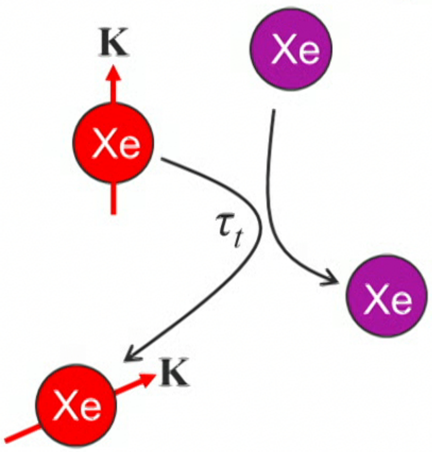
$$\Gamma_{\text{intrinsic}} = \Gamma_t + \Gamma_p$$

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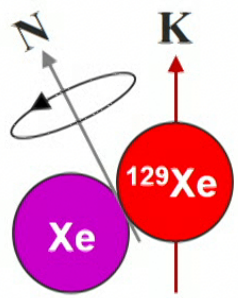
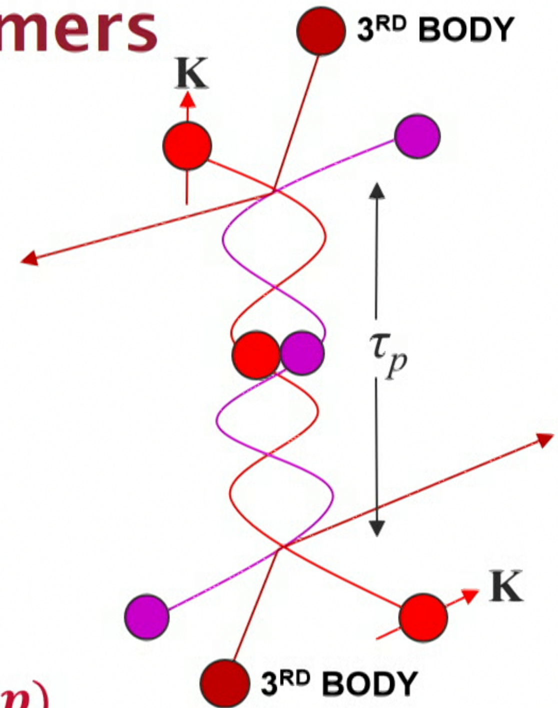



$$C_K(R)\mathbf{N}\cdot\mathbf{K}$$

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* Hunt and Carr (1963); Moudrakovski, et al. (2001)

## PERSISTENT DIMERS ($p$)

- Form/break up in 3-body collisions
- Molecular lifetime $\tau_p \approx 1$ ns ($\approx 10^3\tau_t$!!)
- $\Gamma_p$ independent of [Xe] (for fixed gas composition)

# Persistent-Dimer Relaxation: Theory

$$[\mathrm{Xe}] + [\mathrm{Xe}] \leftrightharpoons [\mathrm{Xe}_2]$$

$$\kappa \equiv \frac{[\mathrm{Xe}_2]}{[\mathrm{Xe}][\mathrm{Xe}]}$$

$$\Gamma_p = \left( \frac{2}{3} \frac{\langle c_K^2 N^2 \rangle}{\hbar^2} \right) \left( \frac{\tau_p}{1 + \omega^2 \tau_p^2} \right) \left( 2\kappa [\mathrm{Xe}] \right)$$

- Mean-squared spin-rotation interaction energy.
- Power spectrum $J(\omega)$ for field fluctuations
  - Lifetime $\tau_p \sim 10^{-9}$ seconds for $[\mathrm{Xe}] \approx 1$ amagat
  - Frequency $\omega/2\pi$ = 11.8 MHz for $B_0$ = 1 T.
  - Can often assume $\omega^2\tau_p^2 << 1$ (fast-fluctuation limit).
- Fraction of atoms bound in molecules, assuming $[\mathrm{Xe}_2] \ll [\mathrm{Xe}]$
  - $[\mathrm{Xe}] \propto \frac{1}{\tau_p}$ ➡ $\Gamma_p$ is *independent* of $[\mathrm{Xe}]$ for $\omega^2\tau_p^2 << 1$.

# Persistent-Dimer Relaxation: Theory

$$[\mathrm{Xe}] + [\mathrm{Xe}] \leftrightarrows [\mathrm{Xe}_2]$$

$$\kappa \equiv \frac{[\mathrm{Xe}_2]}{[\mathrm{Xe}][\mathrm{Xe}]}$$

$$\Gamma_p = \left( \frac{2}{3} \frac{\langle c_K^2 N^2 \rangle}{\hbar^2} \right) \left( \frac{\tau_p}{1 + \omega^2 \tau_p^2} \right) \left( 2\kappa [\mathrm{Xe}] \right)$$

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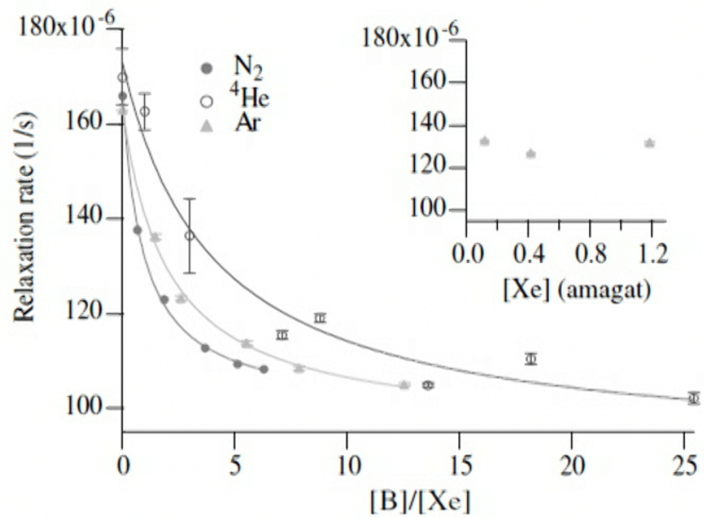
# It *looks* like wall relaxation…

VOLUME 88, NUMBER 11 PHYSICAL REVIEW LETTERS 18 MARCH 2002

## $^{129}$Xe-Xe Molecular Spin Relaxation

B. Chann,[1] I. A. Nelson,[2] L. W. Anderson,[1] B. Driehuys,[2] and T. G. Walker[1]
[1]*Department of Physics, University of Wisconsin–Madison, Madison, Wisconsin 53706*
[2]*Amersham Health, 2500 Meridian Parkway, Suite 175, Durham, North Carolina 27713*
(Received 22 October 2001; published 28 February 2002)



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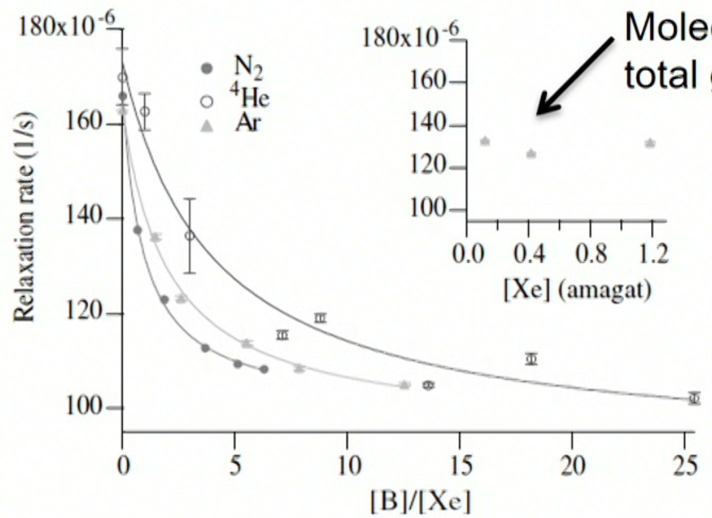
[1]*Department of Physics, University of Wisconsin–Madison, Madison, Wisconsin 53706*
[2]*Amersham Health, 2500 Meridian Parkway, Suite 175, Durham, North Carolina 27713*
(Received 22 October 2001; published 28 February 2002)

…but it's *not* !




Molecular relaxation independent of total gas density for fixed composition.

# Persistent Dimers Usually Dominate $^{129}$Xe Intrinsic Relaxation

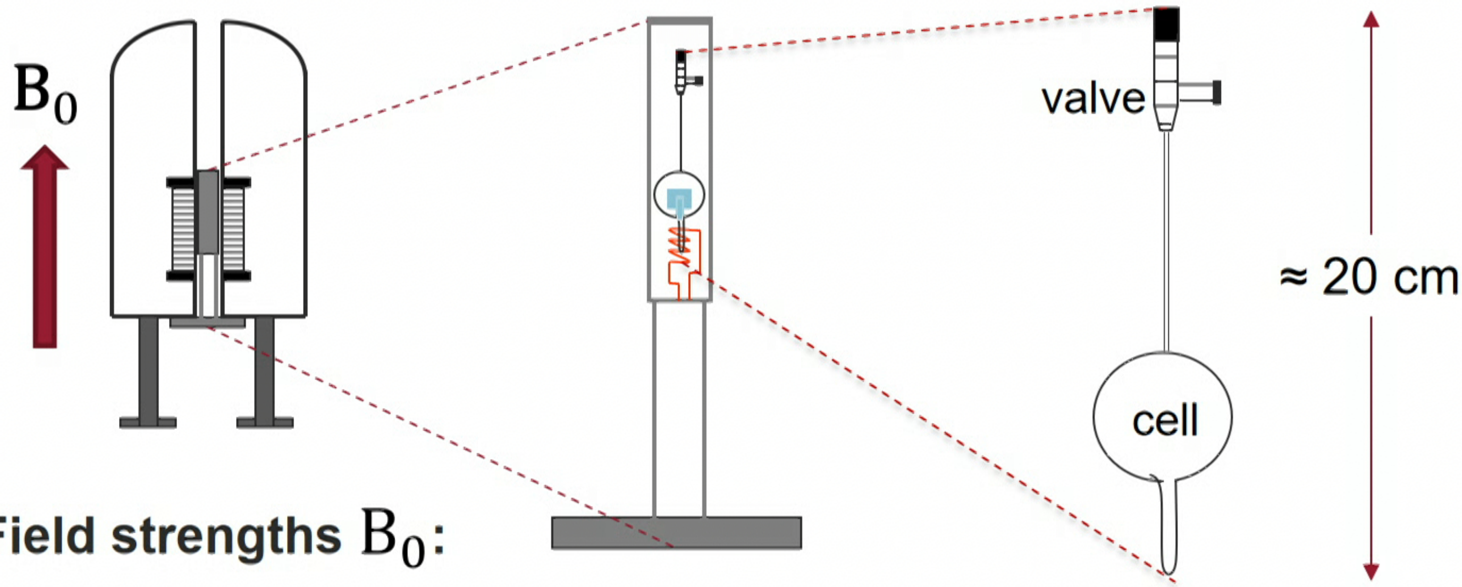
$$[\mathrm{Xe}] + [\mathrm{Xe}] \leftrightharpoons [\mathrm{Xe}_2]$$

$$\kappa \equiv \frac{[\mathrm{Xe}_2]}{[\mathrm{Xe}][\mathrm{Xe}]}$$

$$\Gamma_p = \left( \frac{2}{3} \frac{\left\langle c_K^2 N^2 \right\rangle}{\hbar^2} \right) \left( \frac{\tau_p}{1 + \cancel{\omega^2 \tau_p^2}} \right) \left( 2\kappa [\mathrm{Xe}] \right)$$

- Pure Xe intrinsic relaxation time ≈5 hours
- Second gas acts as a chemical catalyst; decreases $\tau_p$ without changing $\kappa$ ; rate $\Gamma_p$ can be suppressed.

# Shutting Down the Molecules at High-Field




**Field strengths $B_0$:**

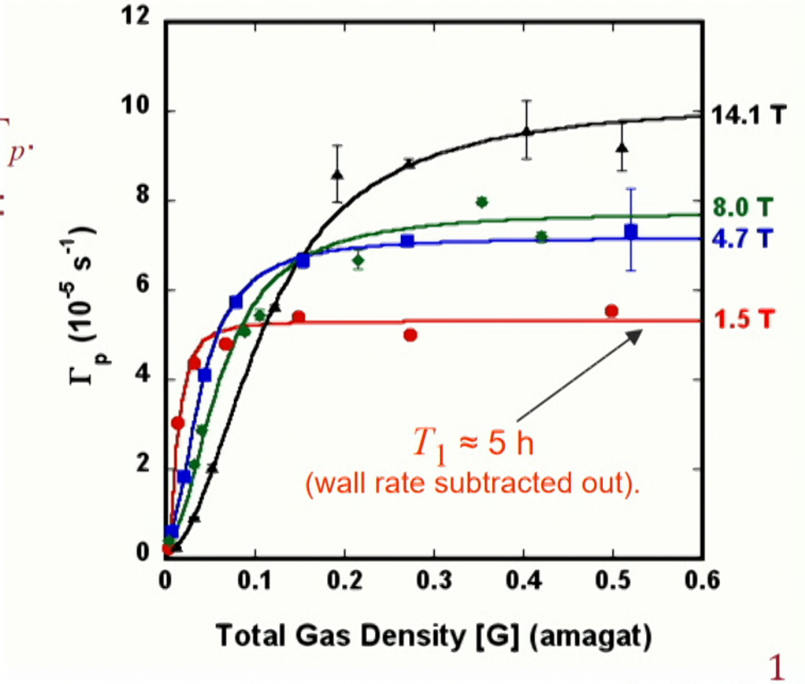
- **1.5 T (17.7 MHz)**
- **4.7 T (55.3 MHz)**
- **8.0 T (94.2 MHz)**
- **14.1 T (166 MHz)**

- **6.7 cm diam spherical cell**
- **Siloxane (SurfaSil) coated**
- **Contains no Rb (HP gas transferred in)**
- **Long and robust wall relaxation times**

# Shutting Down the Molecules at High-Field*

- High gas density: $\Gamma_p$ independent of density.
- Low gas density: magnetic-field suppression of $\Gamma_p$.
- $\Gamma_p$ can make significant contribution to **total** rate:

Limiting $T_1$ **≈ 5 h** for pure Xe
for applied fields $B_0 \leq 1$ T



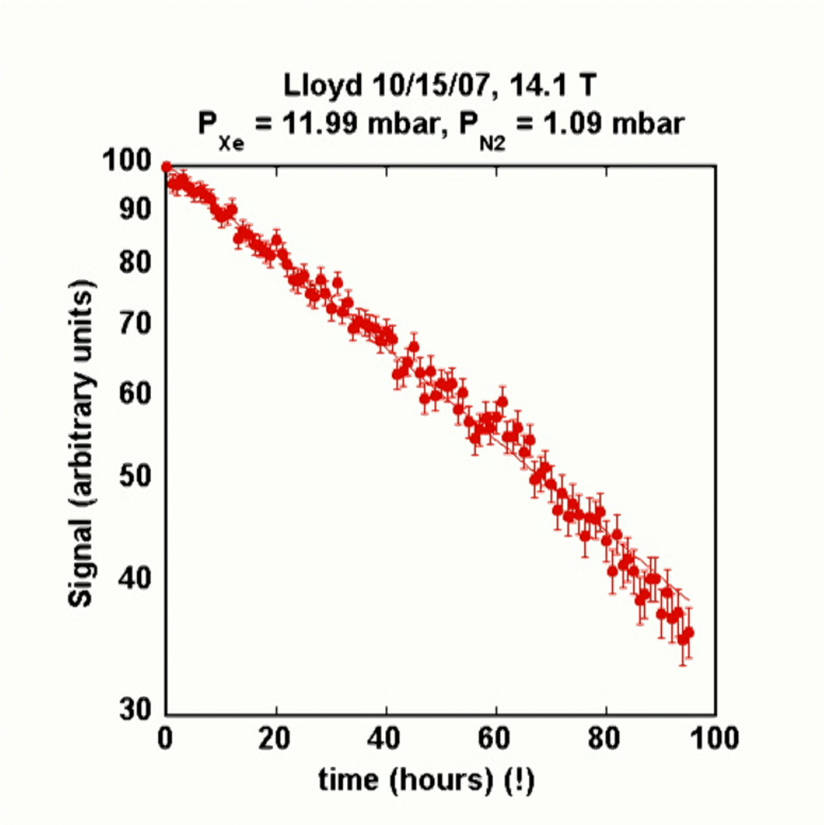

$$\Gamma_p = \left(\frac{2}{3}\frac{\langle c_K^2 N^2\rangle}{\hbar^2}\right)\left(\frac{\tau_p}{1+\omega^2\tau_p^2}\right)(2\kappa[\mathrm{Xe}])$$

$\tau_p \sim \frac{1}{[\mathrm{G}]}$

$[\mathrm{Xe}] \sim [\mathrm{G}]$

*B.N. Berry-Pusey, *et al.*, Phys. Rev. A **74**, 063408 (2006);
B.C. Anger, *et al.*, Phys. Rev. A, Phys. Rev. A **78**, 043406 (2008).

# 100-hour Gas-Phase $T_1$ for $^{129}Xe$



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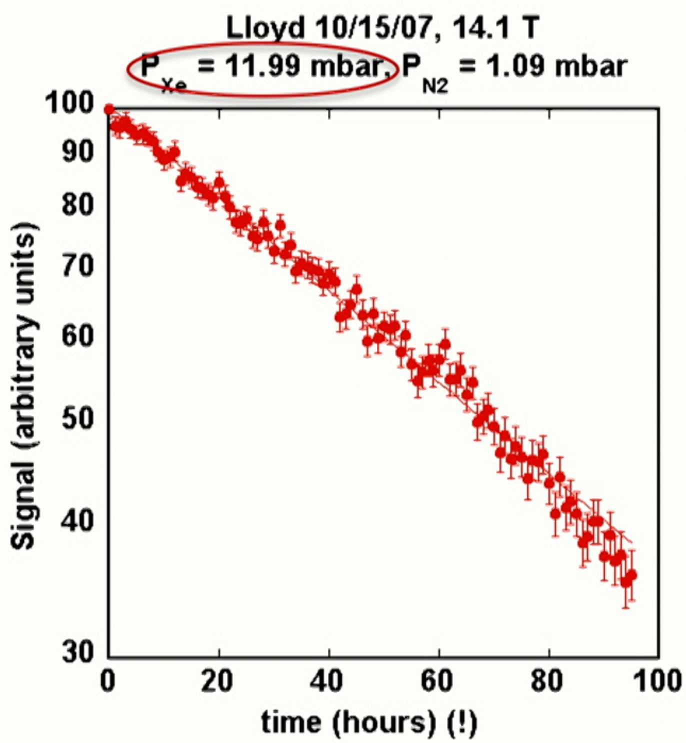
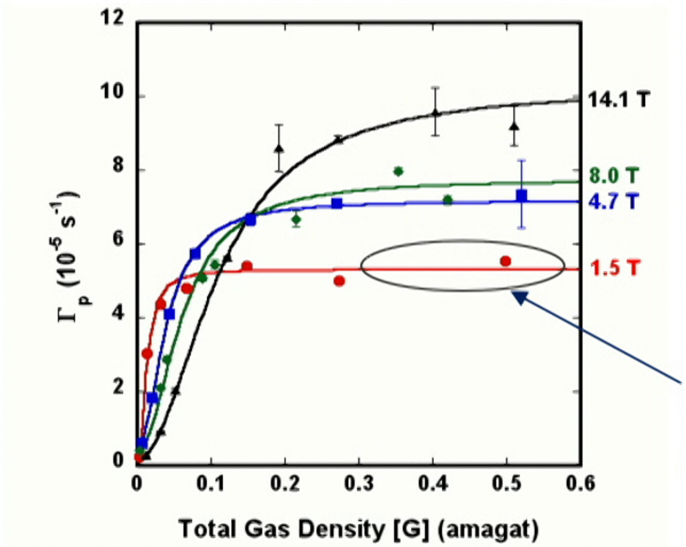
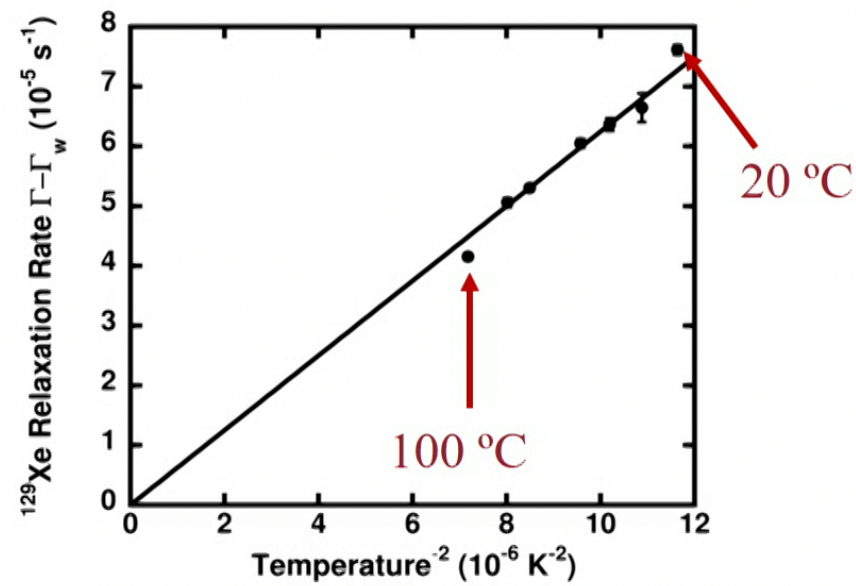



**The Driehuys Axiom**

High $^{129}$Xe polarizations and long $T_1$'s are easy to achieve, provided you don't put any actual xenon in the cell.

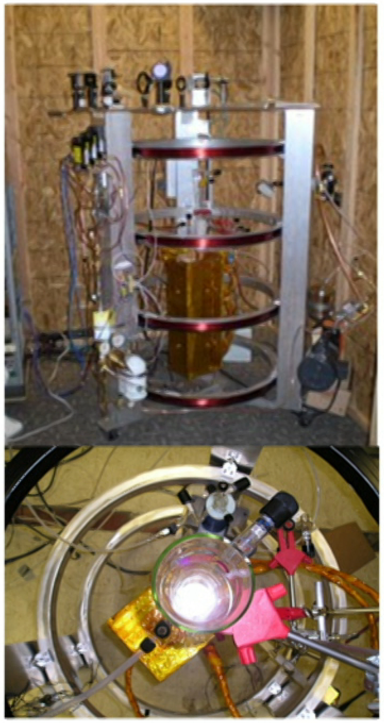
# $Xe_2$ Molecular Relaxation vs. Temperature




- $T_1$ = 5.4 h **measured** in a large 12 cm dia. spherical cell at 30 G, 100 ºC (still limited by wall relaxation)
- Persistent-dimer rate decreases by factor of two going from 20 ºC to 100 ºC; we don't really understand this yet…

# Understanding the Physics of $^{129}$Xe Flow Systems



Hyperpolarized $^{129}$Xe "On Tap"

static "batch" mode $$P_{Xe}(t) = \overline{P_{Rb}}\left(\frac{\gamma_{se}}{\gamma_{se}+\Gamma}\right)\left[1 - e^{-t/\tau_{up}}\right]$$

$$\tau_{up}^{-1} = (\gamma_{se} + \Gamma)$$

flow mode $$P_{Xe}(F) = P_0\left[1 - e^{-F_{crit}/F}\right]$$

Critical flow rate corresponding to a Xe atom spending the optimal time ("spin-up" time) in the cell

# Alkali-Metal Clusters?

PHYSICAL REVIEW A **90**, 023406 (2014)

**Characterizing and modeling the efficiency limits in large-scale production of hyperpolarized $^{129}$Xe**

M. S. Freeman,[1,2] K. Emami,[3] and B. Driehuys[1,2,*]
[1] *Center for In Vivo Microscopy, Department of Radiology, Duke University, 311 Research Drive, Durham, North Carolina 27710, USA*
[2] *Medical Physics Graduate Program, Duke University, 2424 Erwin Road, Durham, North Carolina 27710, USA*
[3] *Polarean, Inc., 2500 Meridian Pkwy #175, Durham, North Carolina 27713, USA*
(Received 17 February 2014; revised manuscript received 22 June 2014; published 6 August 2014)

The European Physical Journal D
May 2012, 66:140

Explosive evaporation of Rb or K fractal clusters by low power CW radiation in the presence of excited atoms

Authors | Authors and affiliations

S. N. Atutov, A. I. Plekhanov, A. M. Shalagin, R. Calabrese, L. Tomassetti, V. Guidi

# PiNG 2017

http://www.physics.utah.edu/ping

## Conference on Polarization in Noble Gases

OCTOBER 8 – 13, 2017; PARK CITY, UTAH, USA

TOPICS:

- Basics of Optical Pumping (MEOP & SEOP)
- Polarization hardware, lasers, new techniques
- MR Imaging with hyperpolarized gases
- Magnetometry
- $^{3}$He Neutron Spin Filters
- Polarized targets
- Porous Media/Surface Science
- Fundamental Symmetries Tests
- $^{129}$Xe Biosensors/HyperCEST

INVITED SPEAKERS:

- Mitchell Albert *Thunder Bay RRI*
- Talissa A. Altes *University of Missouri*
- Gordon D. Cates *University of Virginia*
- Wangchun Chen *NIST-Gaithersburg*
- Boyd Goodson *Southern Illinois University*
- G. Wilson Miller *University of Virginia*
- Graham Norquay *University of Sheffield*
- Pierre-Jean Nacher *LKB-Paris*
- W. Michael Snow, *Indiana University*
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# Thank you!