

QUALIFIER

Mark Limes April 1st, 2011





Introduction

- Outline of xenon storage cell project
- Optical pumping
- Brief interlude
- Gas-phase spin-exchange
- Relaxation
- Low-field nuclear magnetic resonance
- Preliminary results
- Discussion of results
- Current and future projects

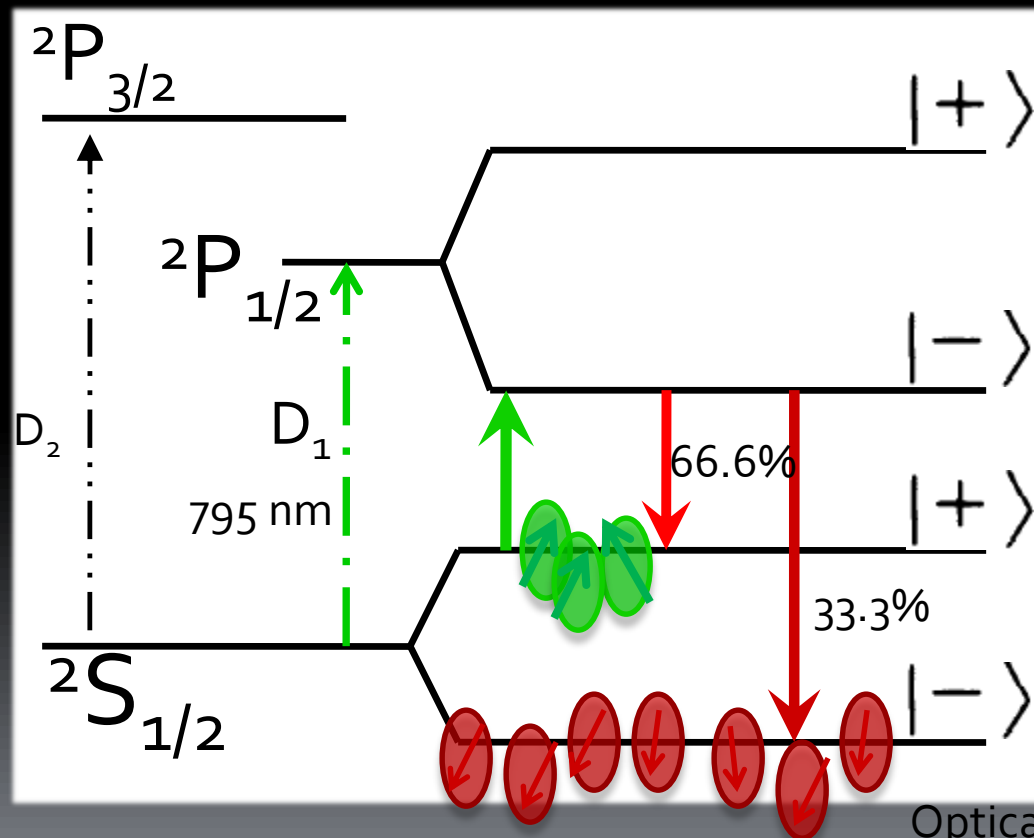
Project Purpose

- Find a holding method for (hyperpolarized) xenon that minimizes nuclear polarization relaxation rates
- Develop a consistent system of characterizing the quality of holding methods
- Collect data on and provide explanations for various intrinsic and extrinsic physical mechanisms causing relaxation



Optical Pumping

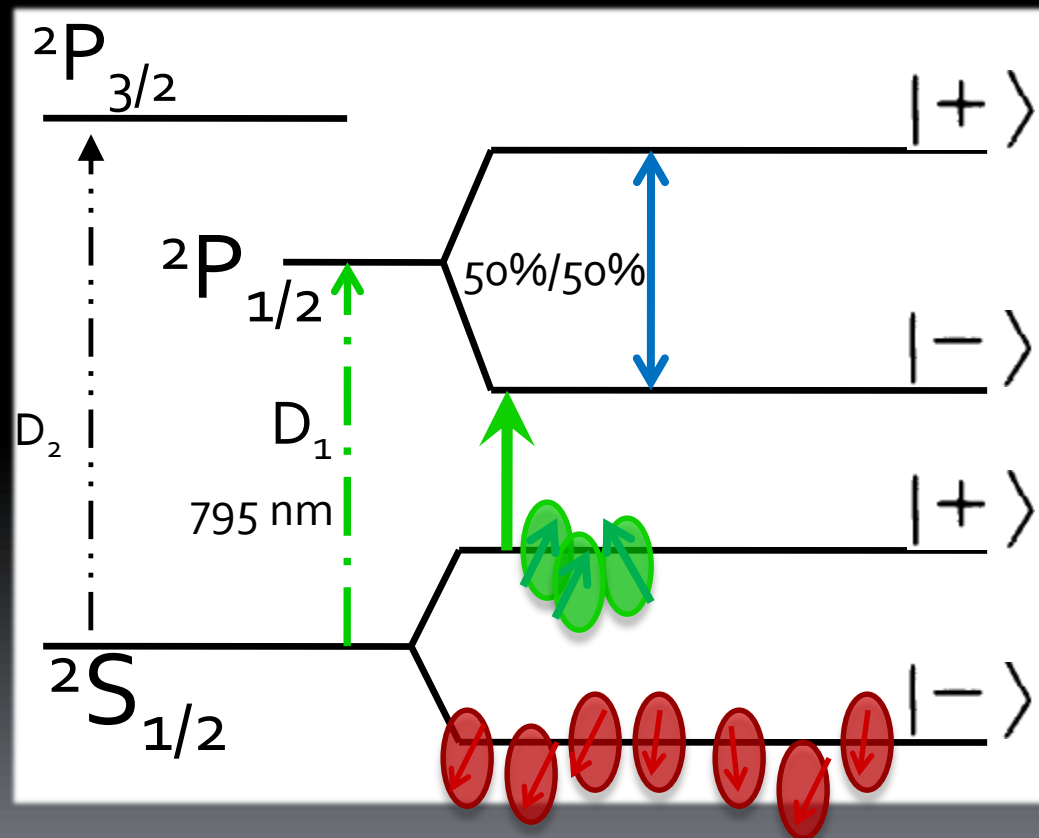
- Optically pumping an alkali-metal's valence electron state with circularly polarized light



Optically Pumped Atoms-
Happer, 2010

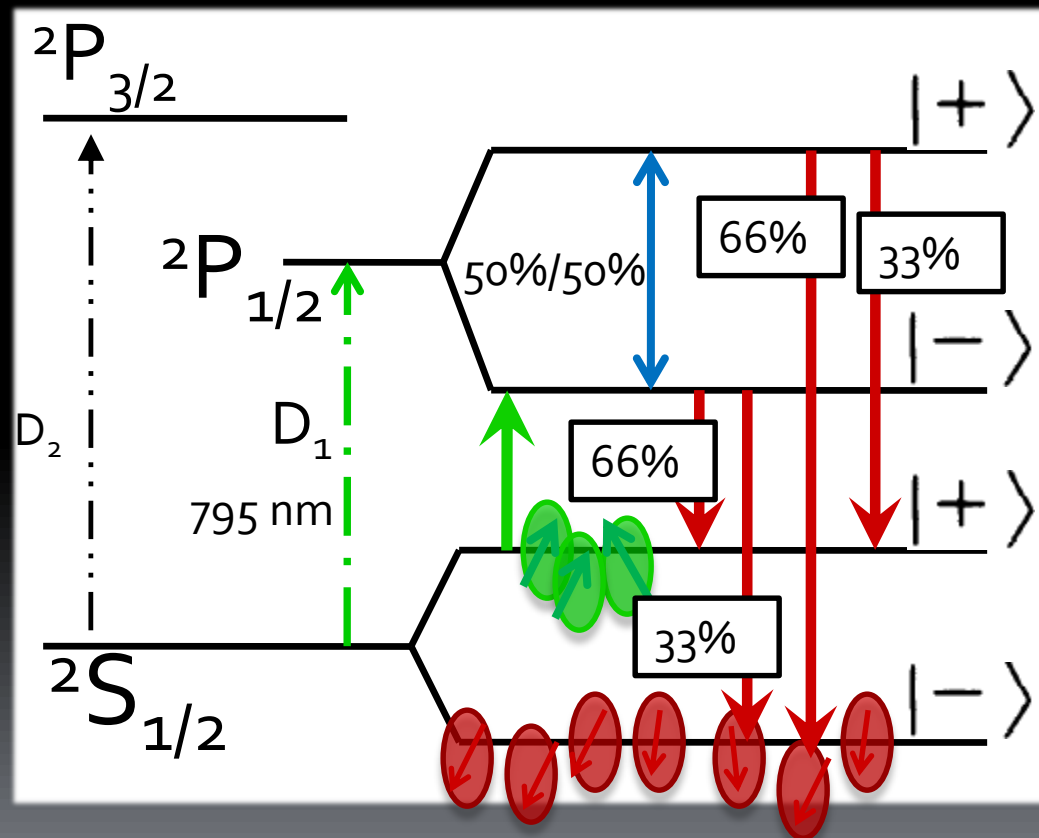
Optical Pumping

- Buffer gases depolarize the excited state and increase photon efficiency by collisional mixing



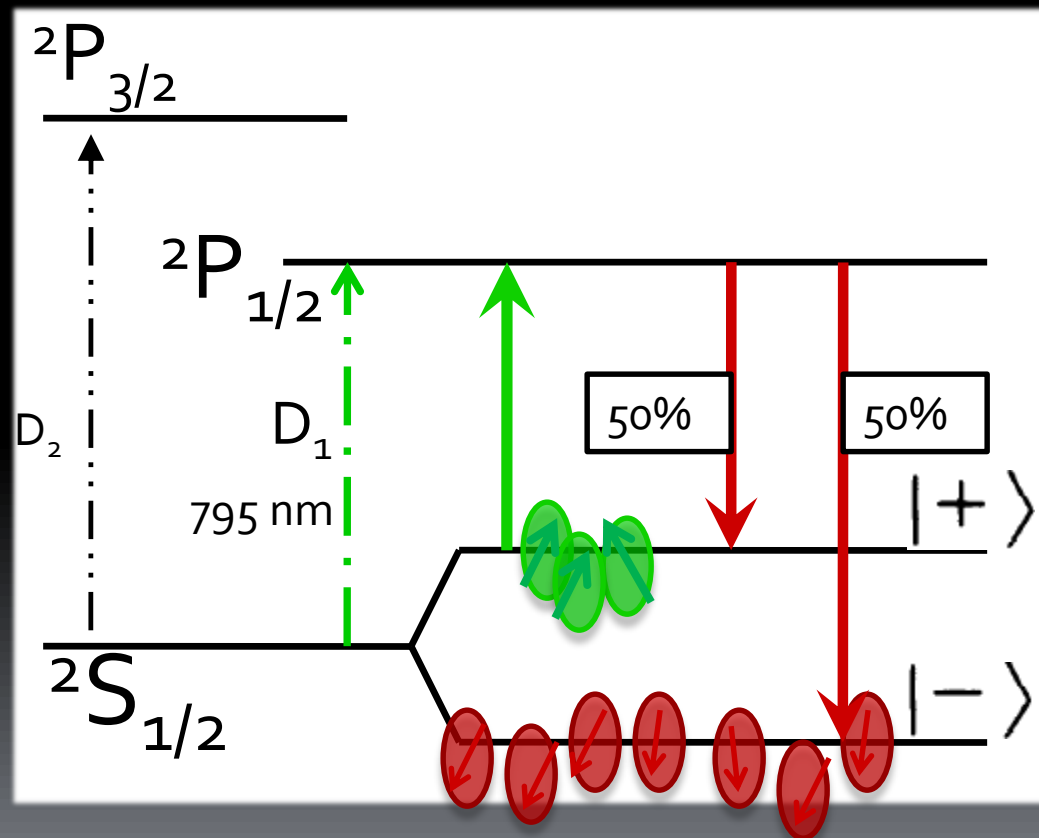
Optical Pumping

- Buffer gases depolarize the excited state and increase photon efficiency by collisional mixing



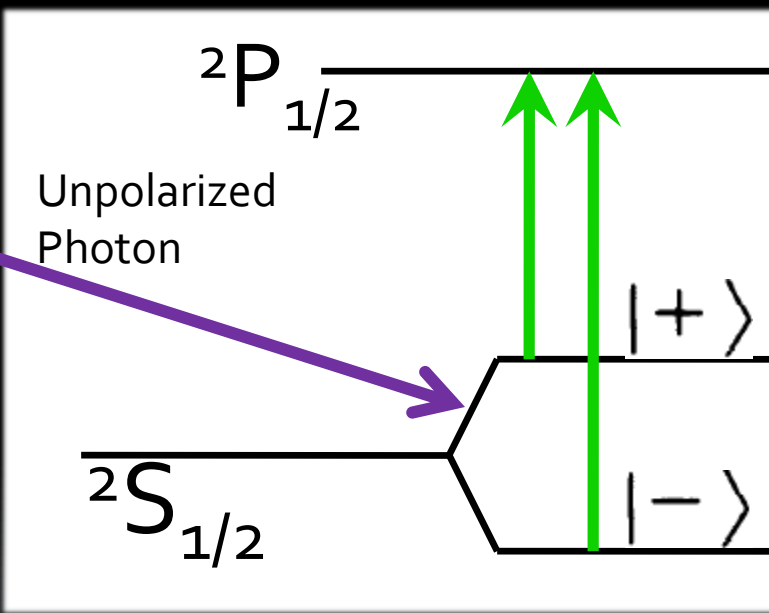
Optical Pumping

- Buffer gases depolarize the excited state and increase photon efficiency by collisional mixing



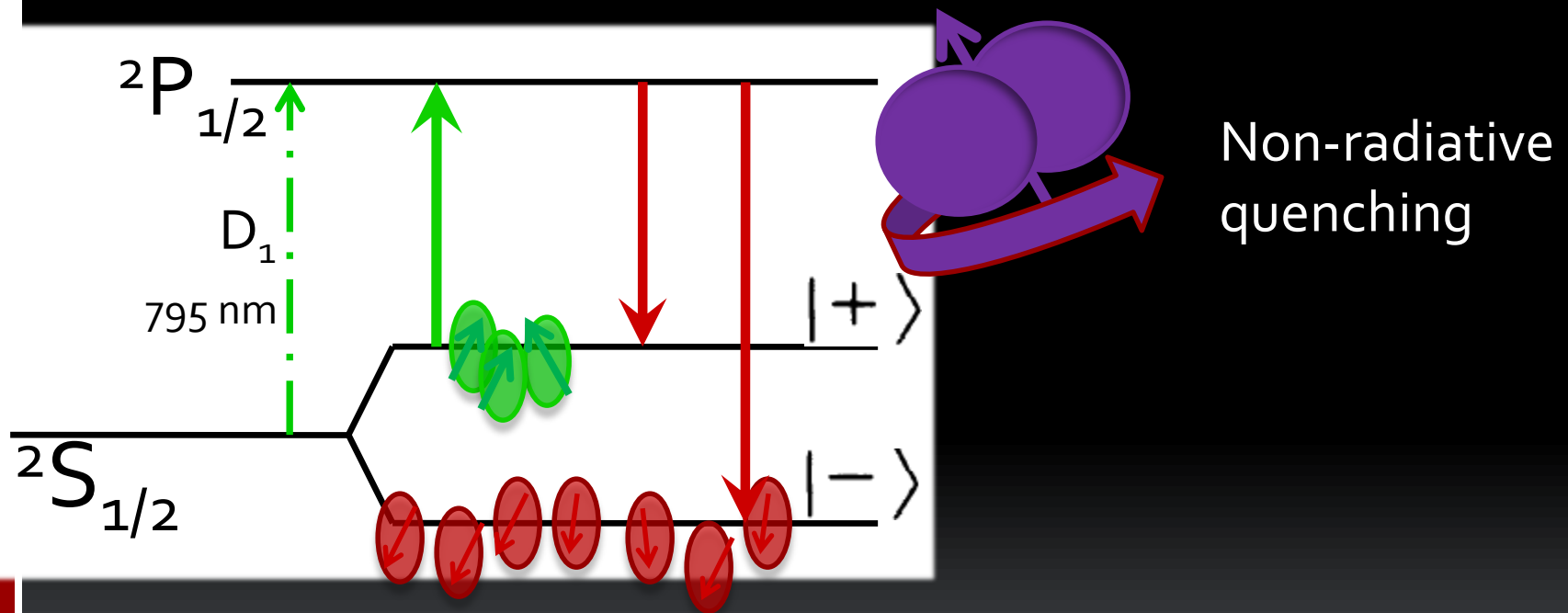
SECRET

- 100



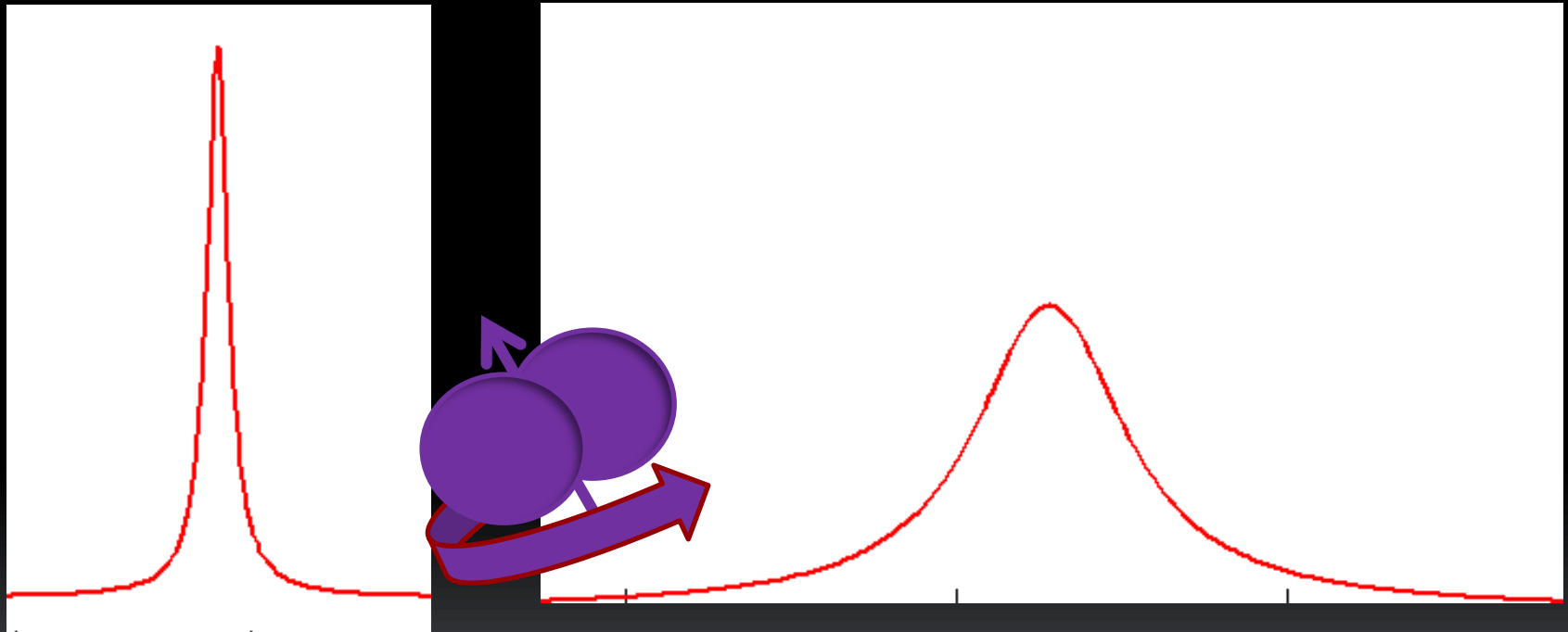
Optical Pumping

- Buffer gas prevents radiation trapping and takes away angular momentum



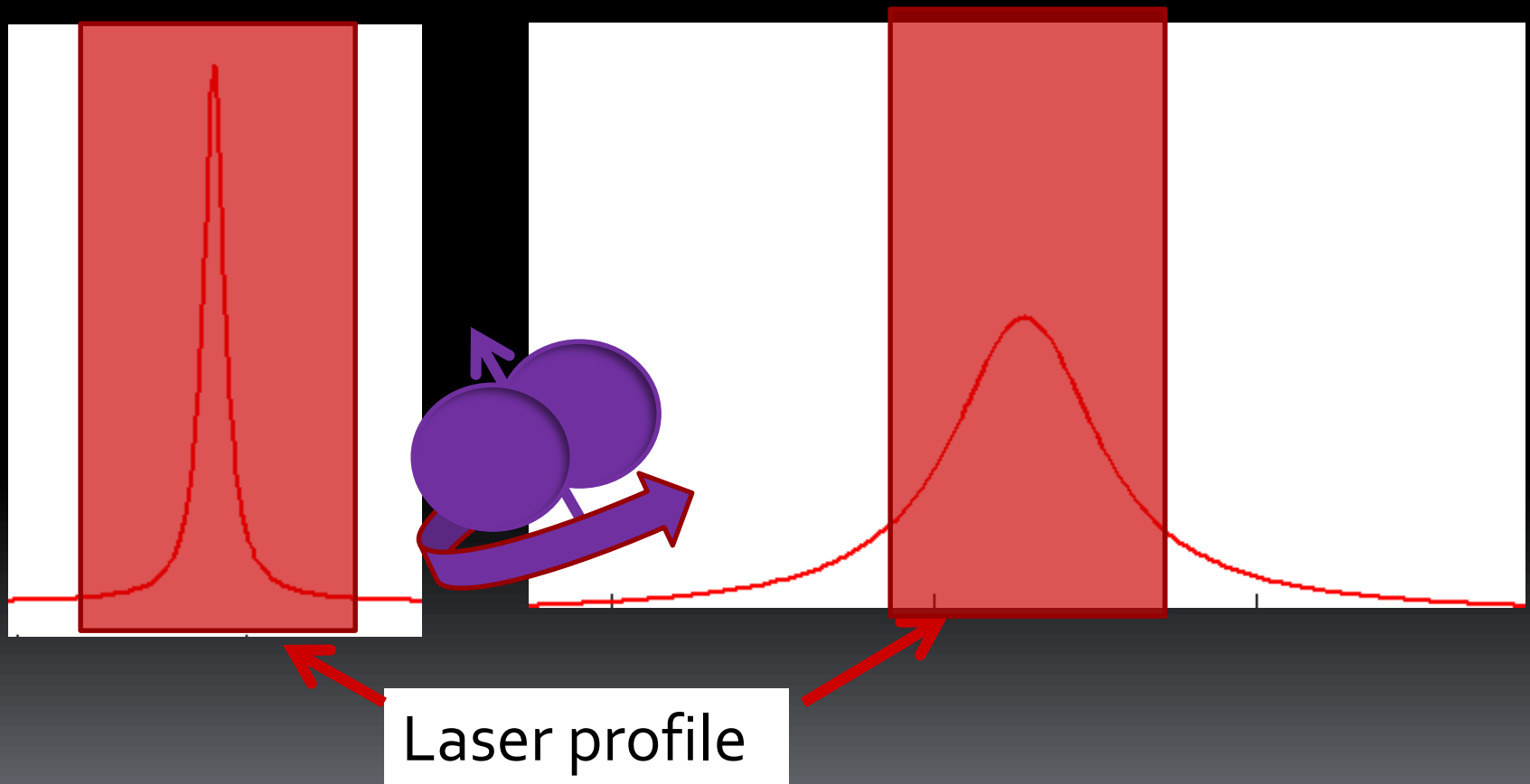
Optical Pumping

- Buffer gas also broadens absorption line



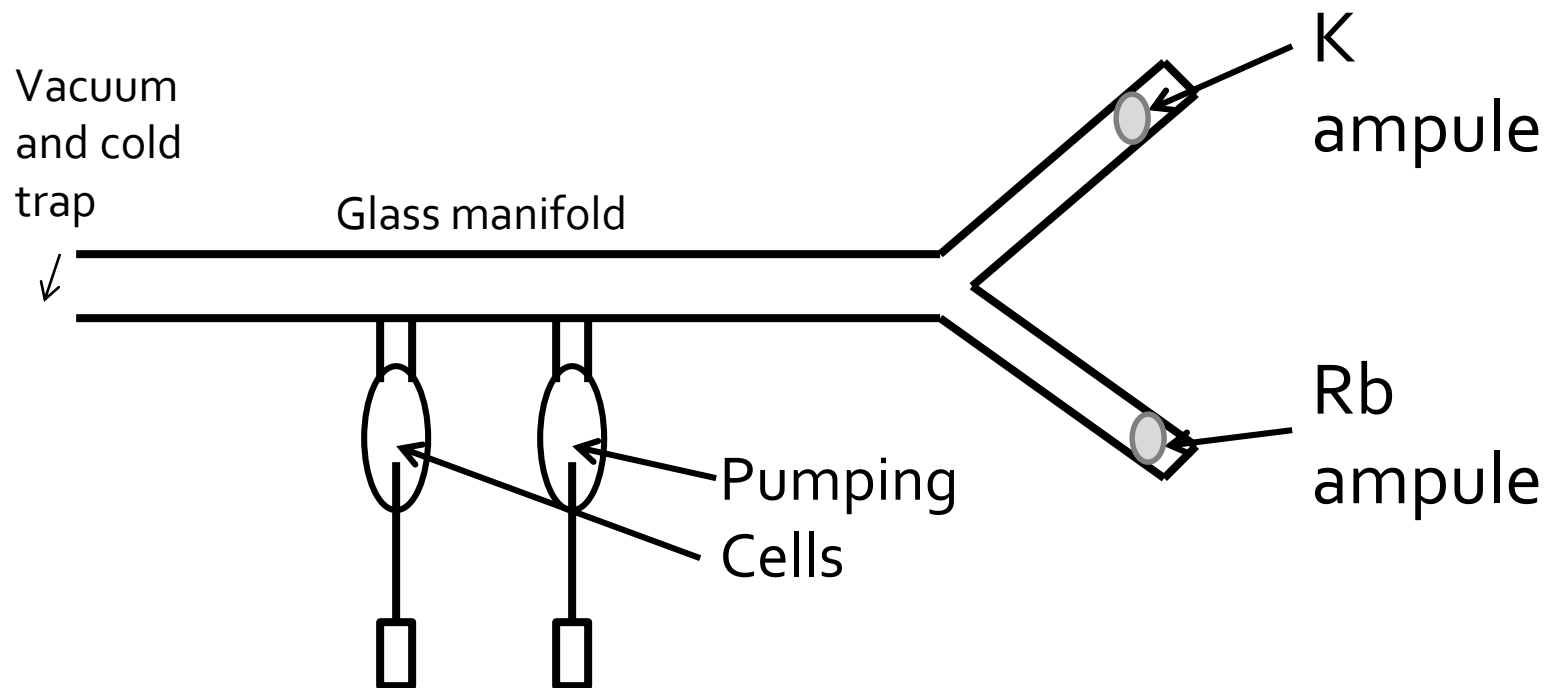
Optical Pumping

- Buffer gas also broadens absorption line



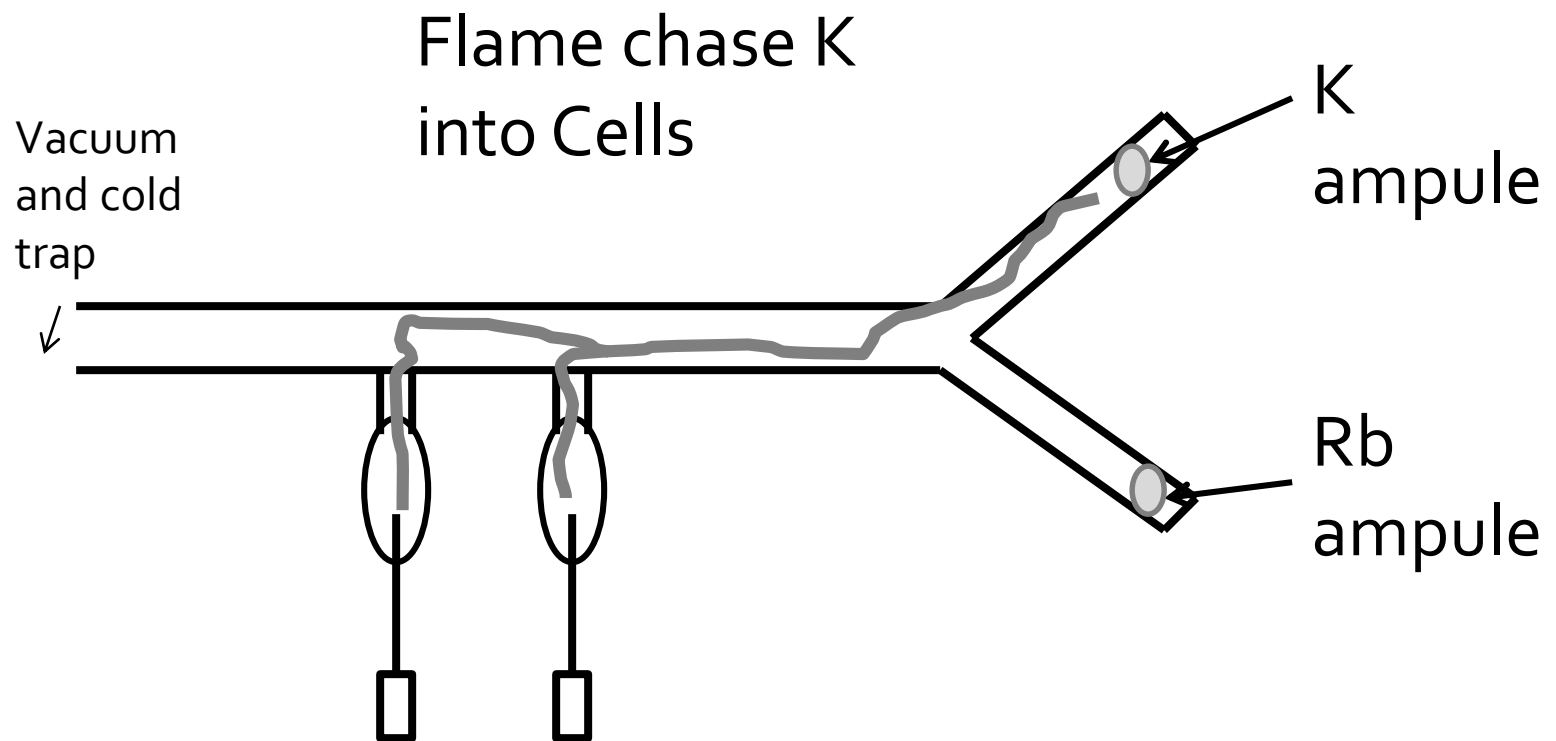
Brief Interlude

- Hybrid K-Rb cells



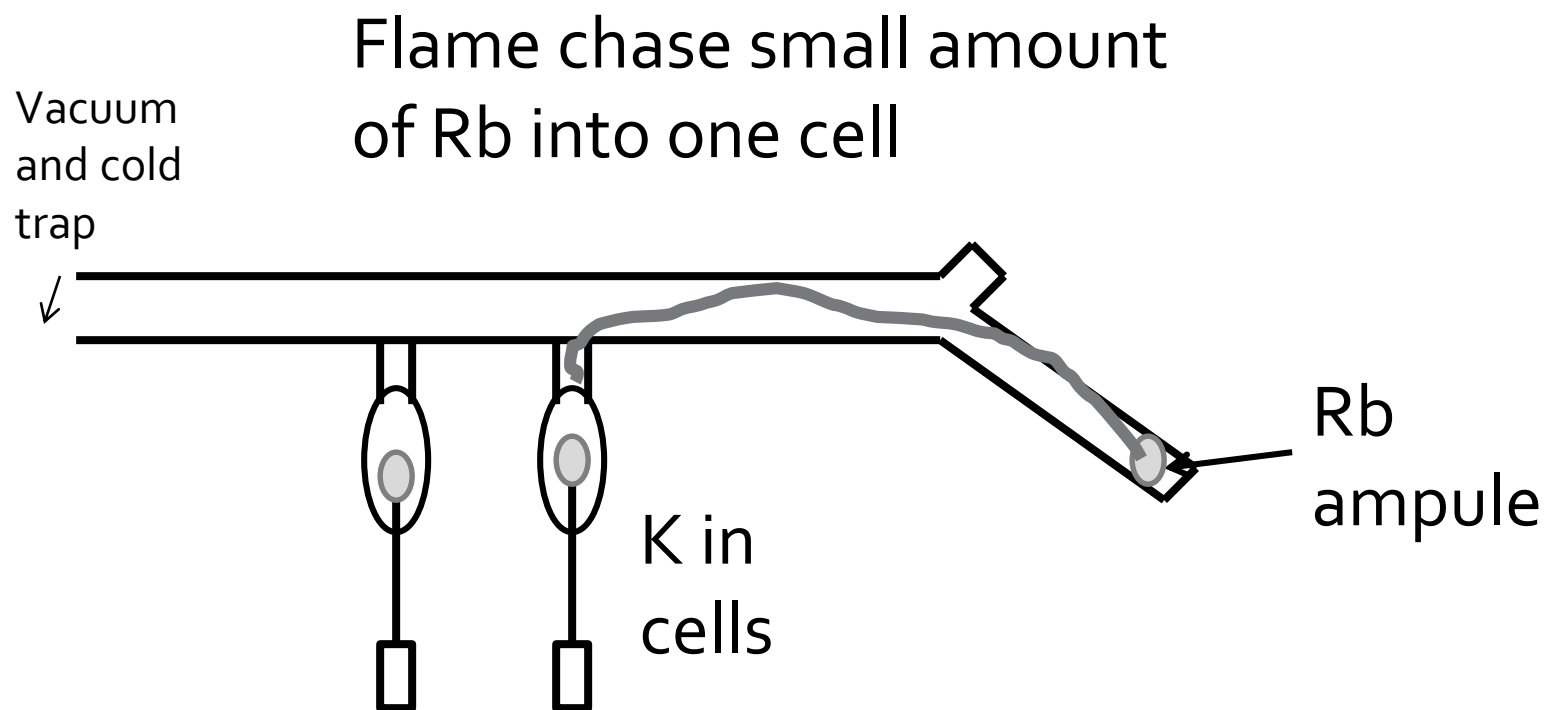
Brief Interlude

- Hybrid K-Rb cells



Brief Interlude

- Hybrid K-Rb cells

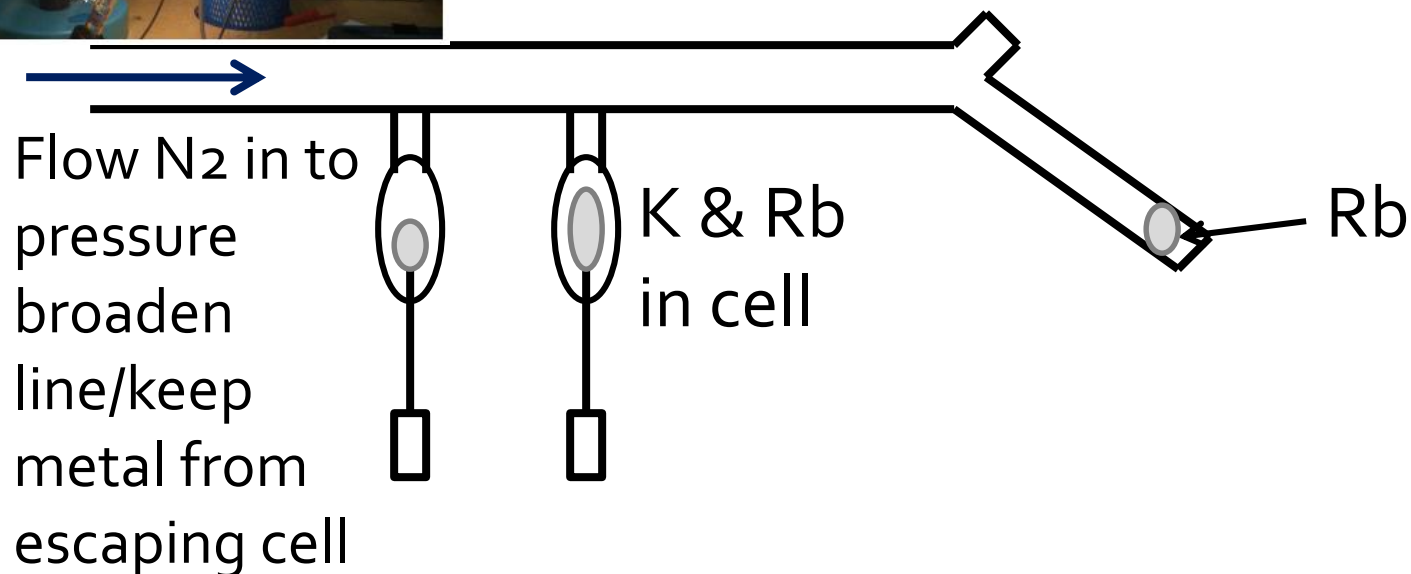


Brief Interlude

- Hybrid K-Rb cells



Run a white light absorption measurement, analyze, able to add more Rb



Brief Interlude

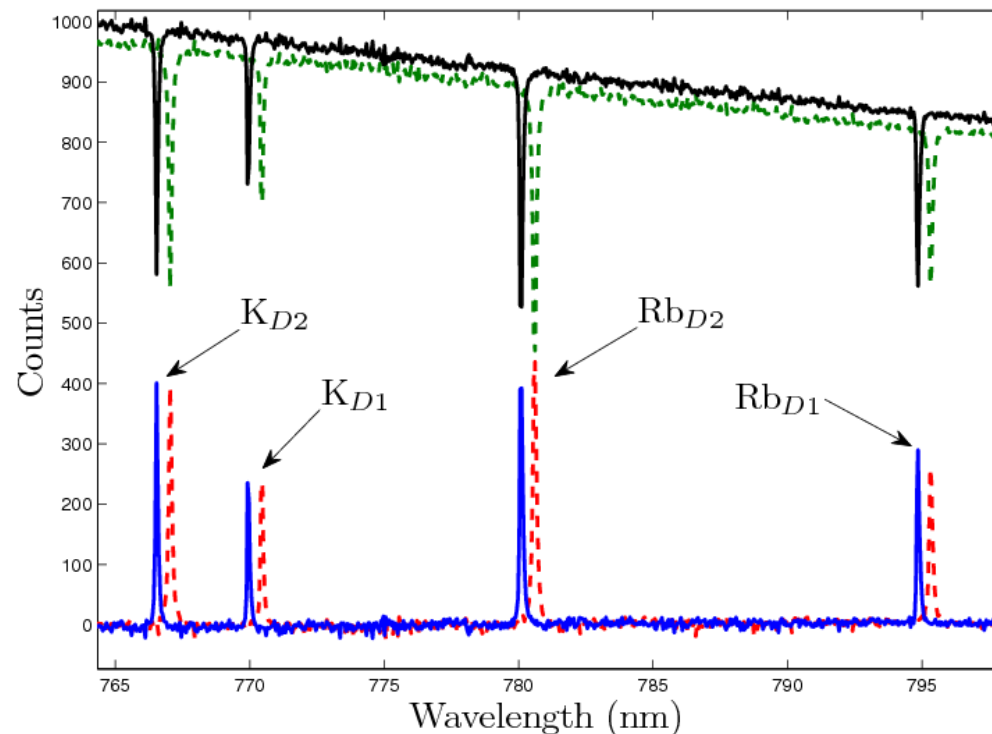
Hybrid K-Rb cells

154B in situ at 105C

| | Lorentzian |
|-----------------------|----------------|
| K_{D1}/K_{D2} | $.64 \pm .04$ |
| Rb_{D1}/Rb_{D2} | $.62 \pm .04$ |
| D_{105C} (D2 lines) | $.695 \pm .03$ |
| D_{105C} (D1 lines) | $.719 \pm .05$ |
| D_{200C} (D2 lines) | $1.15 \pm .05$ |
| D_{200C} (D1 lines) | $1.19 \pm .09$ |

154B Post pull-off at 100C

| | Lorentzian |
|-----------------------|----------------|
| K_{D1}/K_{D2} | $.52 \pm .02$ |
| Rb_{D1}/Rb_{D2} | $.55 \pm .02$ |
| D_{100C} (D2 lines) | $.757 \pm .02$ |
| D_{100C} (D1 lines) | $.726 \pm .03$ |
| D_{200C} (D2 lines) | $1.30 \pm .03$ |
| D_{200C} (D1 lines) | $1.25 \pm .04$ |

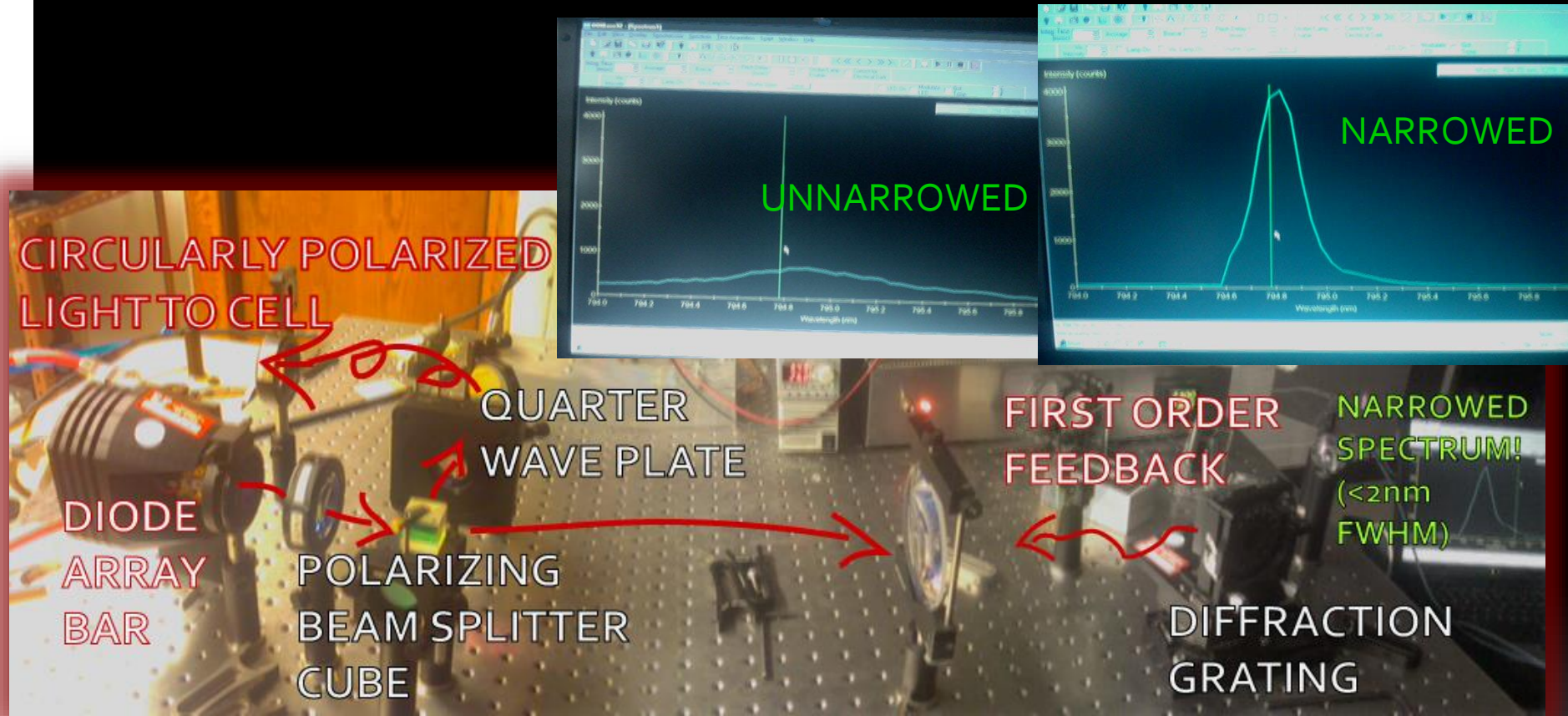


Solid lines – in situ

Dash lines – post-pull off (shifted for clarity)

Raw data on top, bottom baseline corrected

Optical Pumping



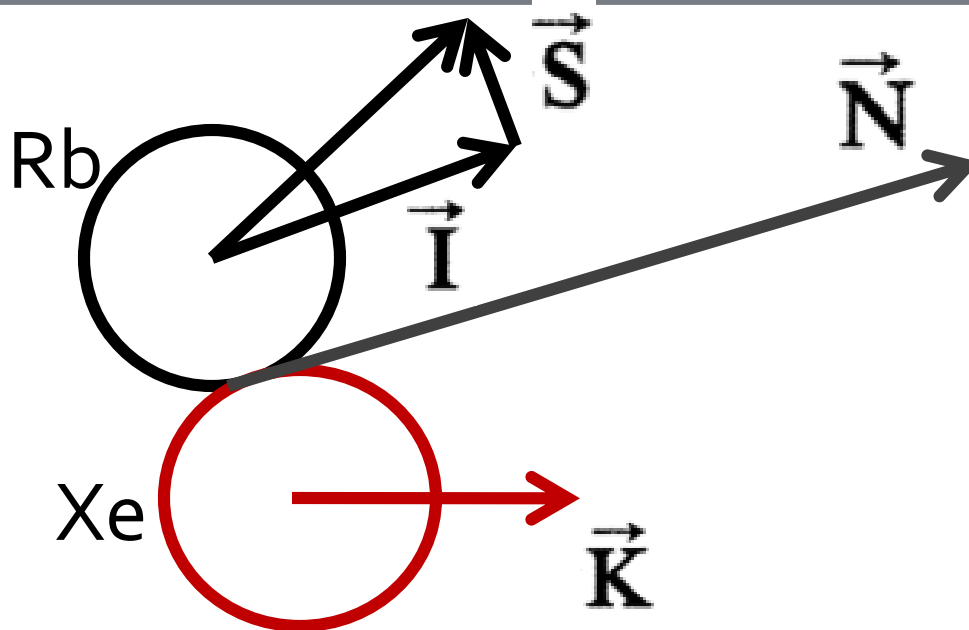
- One of our laser set-ups

(Chann et. al. 2000,

Gas-Phase Spin-Exchange

- Hamiltonian

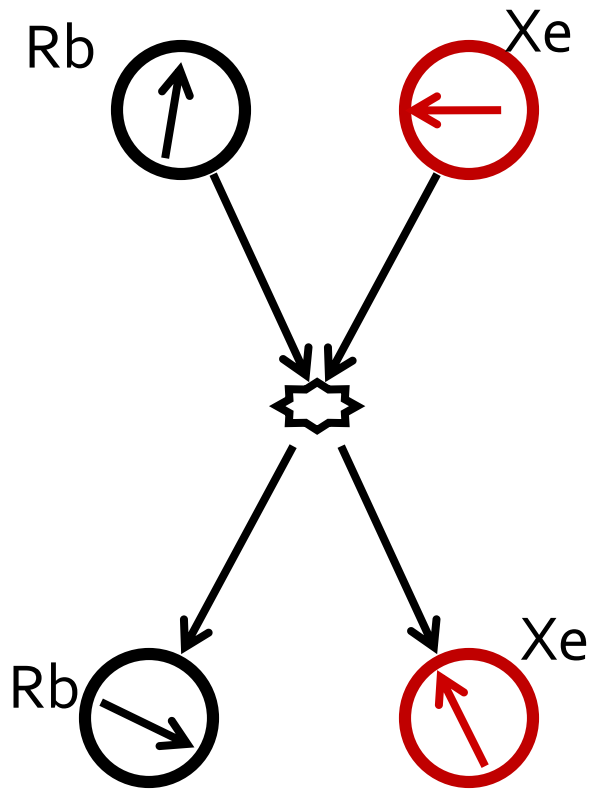
$$H = A \vec{I} \cdot \vec{S} + \gamma \vec{N} \cdot \vec{S} + \alpha \vec{K} \cdot \vec{S} + \dots$$



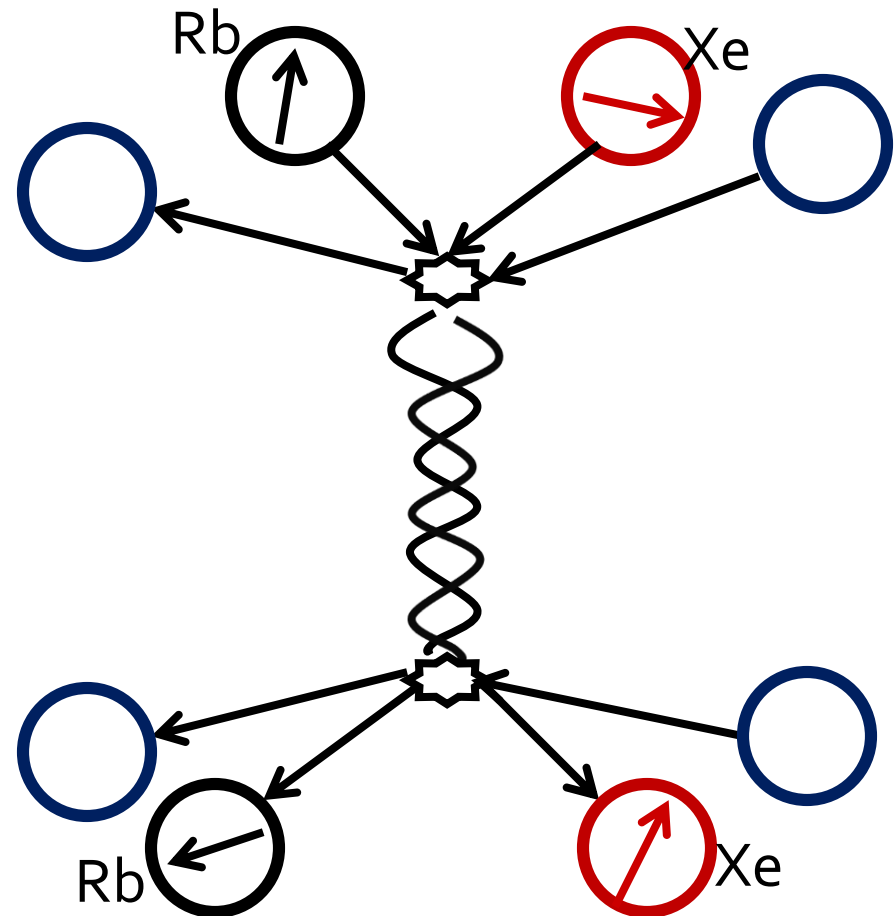
Phys. Rev. A Happer 1984

Gas-Phase Spin-Exchange

- Binary collisions and Van der Waals molecules



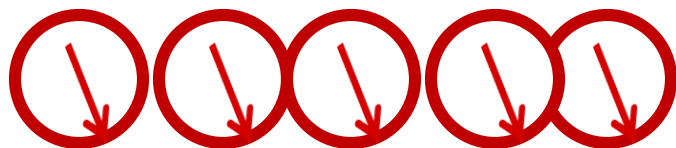
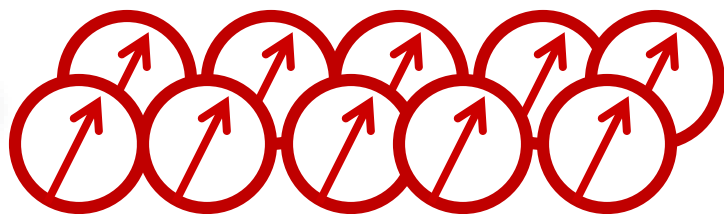
Phys. Rev. A Happer 1984



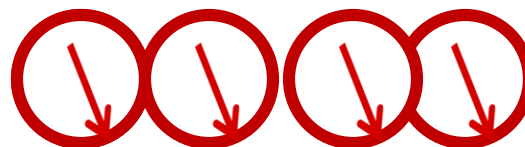
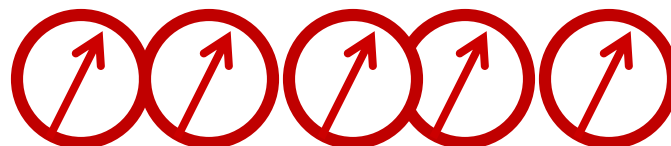
Gas-Phase Spin-Exchange

- Why go to all this trouble?

HYPERPOLARIZED

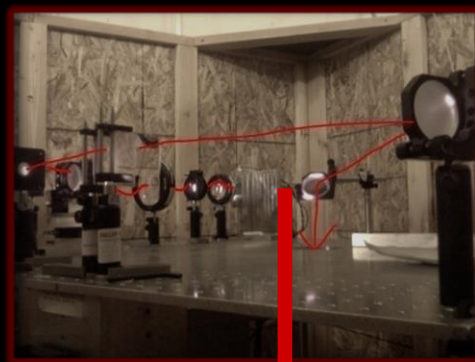


THERMAL



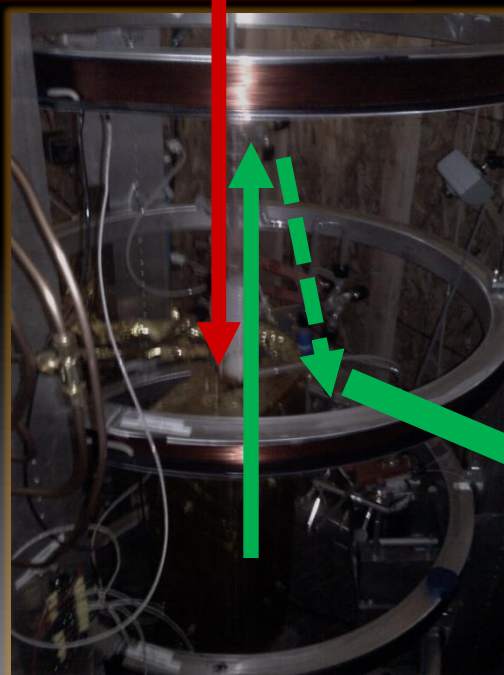
Polarizer

- Flow through Xenon polarizer – a useful tool



Rb
Pump

Flow Xe



COLLECT
XENON



Relaxation

- Longitudinal Nuclear Spin Relaxation

$$\frac{dn}{dt} = \frac{n_o - n}{T_1}$$

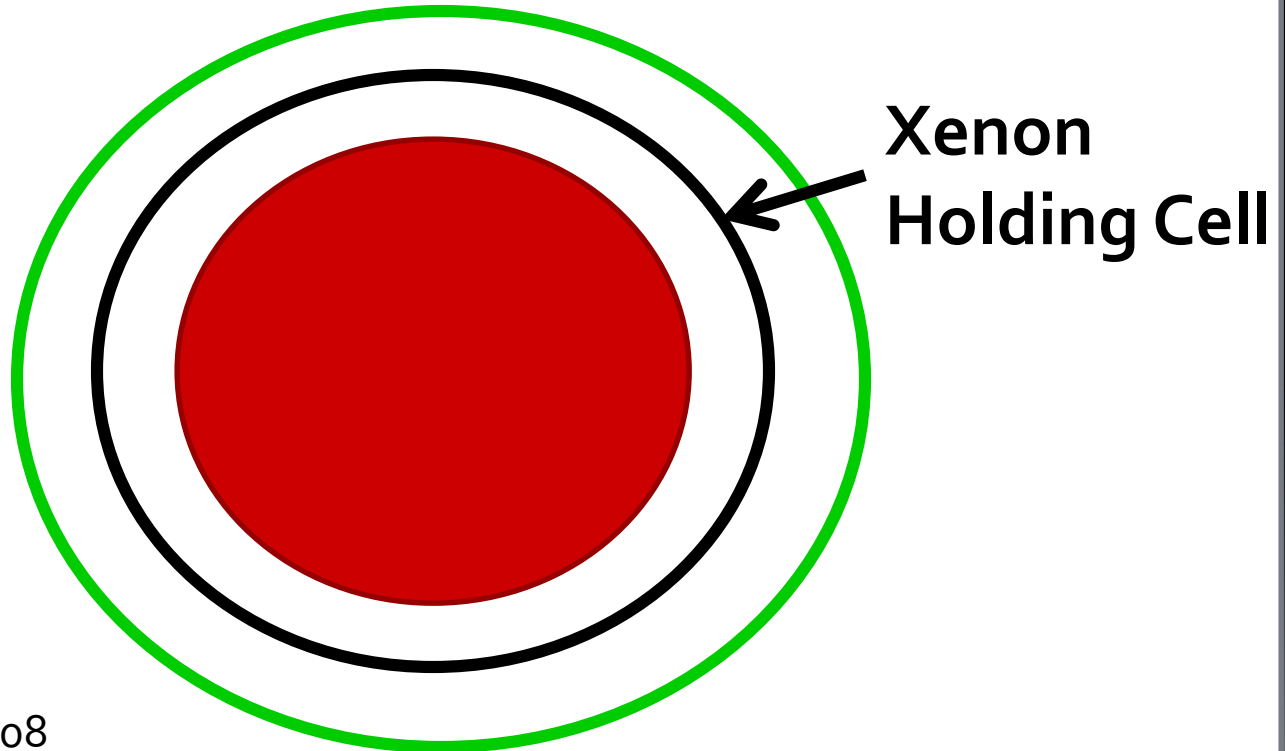
$$n = Ae^{-t/T_1}$$

where n is population difference

Relaxation

- Intrinsic and extrinsic

$$\Gamma = \Gamma_t + \Gamma_p + \Gamma_g + \Gamma_w$$

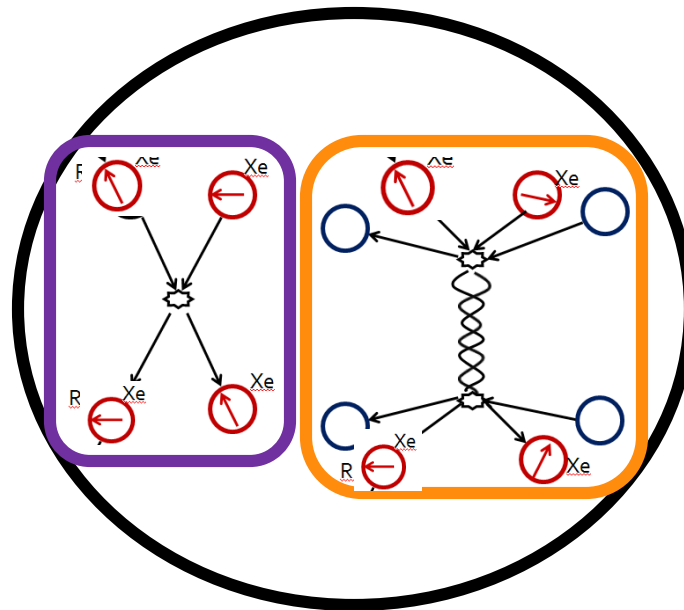


Phys. Rev. A-
Anger et. al. 2008

Relaxation

- Intrinsic: Transient and persistent

$$\Gamma = \Gamma_t + \Gamma_p + \Gamma_g + \Gamma_w$$

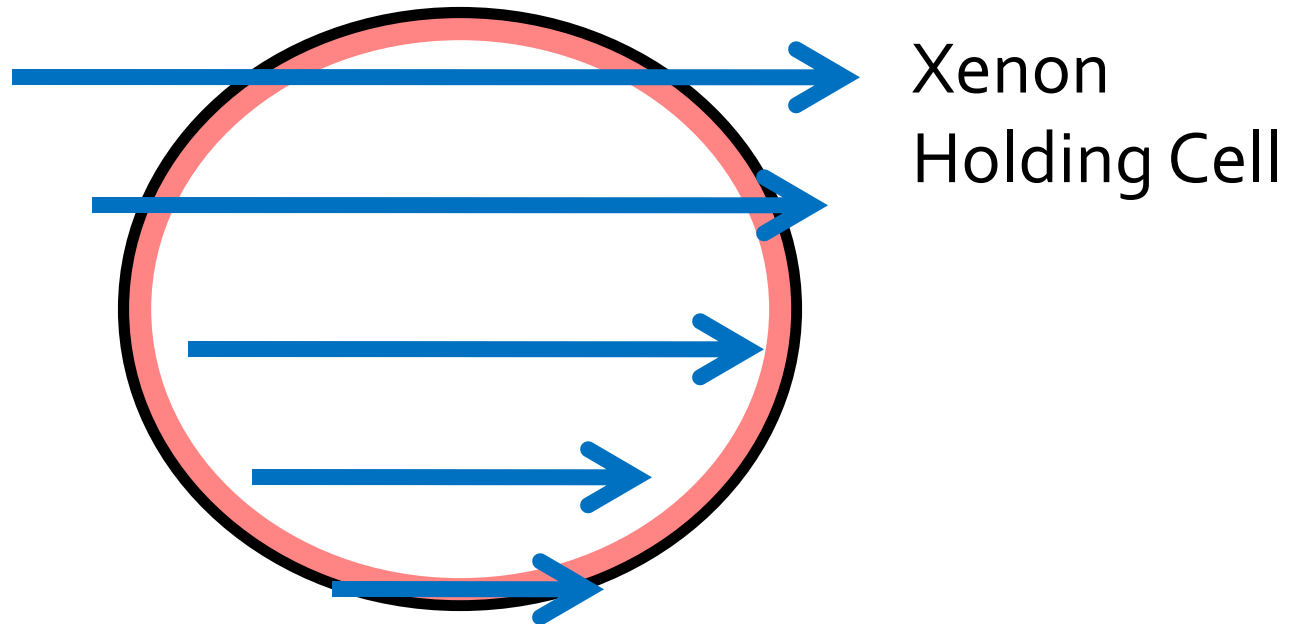


Xenon
Holding Cell

Relaxation

- **Extrinsic:** Magnetic field gradient and wall

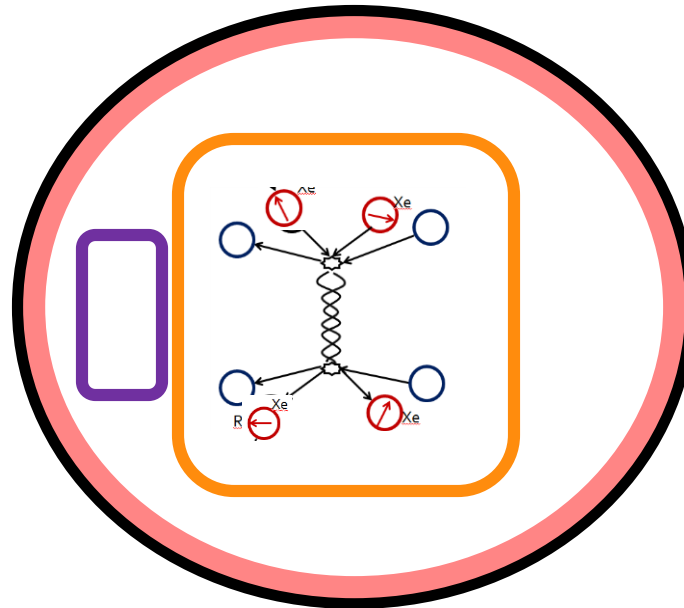
$$\Gamma = \Gamma_t + \Gamma_p + \Gamma_g + \Gamma_w$$



Relaxation

- This work is in the regime primarily dominated by **persistent dimers** and **wall**

$$\Gamma = \Gamma_t + \Gamma_p + \Gamma_g + \Gamma_w$$



Xenon
Holding Cell

Relaxation

- Semi-phenomenological equation for xenon nuclear spin relaxation rate

$$\Gamma_{-} = \frac{[\text{Xe}]}{56.1 \text{ h}} \left(\frac{T_o}{T} \right)^{\frac{1}{2}} + \frac{1}{4.59 \text{ h}} \left(1 + r \frac{[B]}{[\text{Xe}]} \right)^{-1} \left(\frac{T_o}{T} \right)^2 + \Gamma_w$$

Persistent dimer term

Wall term

Transient dimer term

Gradient term ignored

Phys. Rev. A
Anger et. al. 2008

Relaxation

- Buffer gas helps yet again

$$\Gamma_p \propto \tau_B$$

$$\frac{1}{\tau_B} = k_{\mathbf{B}}[\mathbf{B}] + k_{\mathbf{Xe}}[\mathbf{Xe}]$$

$$\Gamma_p = \frac{\Gamma_p^{\mathbf{Xe}}}{1 + r \frac{[\mathbf{B}]}{[\mathbf{Xe}]}}$$

$$\text{where } r = \frac{k_{\mathbf{B}}}{k_{\mathbf{Xe}}}$$

Relaxation

- Unexpected temperature dependence

$$\Gamma_p = 2\mathcal{K}[\text{Xe}] \frac{W}{\tau_B}$$

$$\Gamma_p \propto T^{\frac{1}{2}} \tau_B$$

$$\tau_B = (\bar{V} \sigma_o n)^{-1} \propto T^{-\frac{1}{2}} \sigma_o^{-\frac{1}{2}}$$

$$\Gamma_p \propto \sigma_o^{-\frac{1}{2}}$$

where W is fraction of polarization lost during lifetime τ_B of the molecule, \mathcal{K} is the chemical equilibrium coefficient, \bar{V} is the mean relative velocity, σ_o is the mean total cross section at this speed, and n is the mean number of molecules per unit volume.

Phys. Rev. A
Anger et. al. 2008

Phys. Rev. Lett.
Chann et. al. 2002

Phys. Rev. A
Walker et. al. 2001

Relaxation

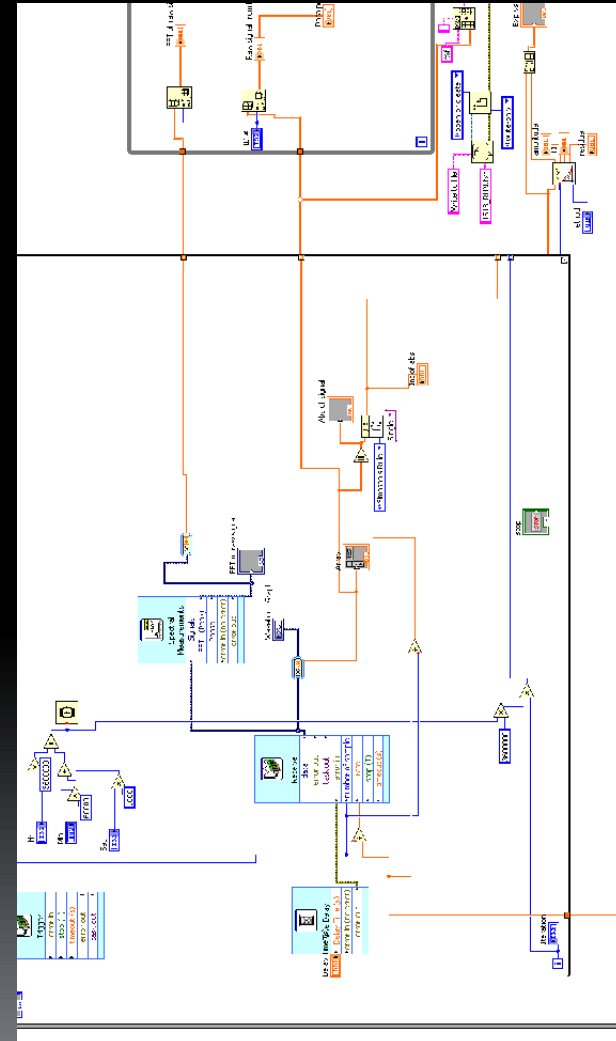
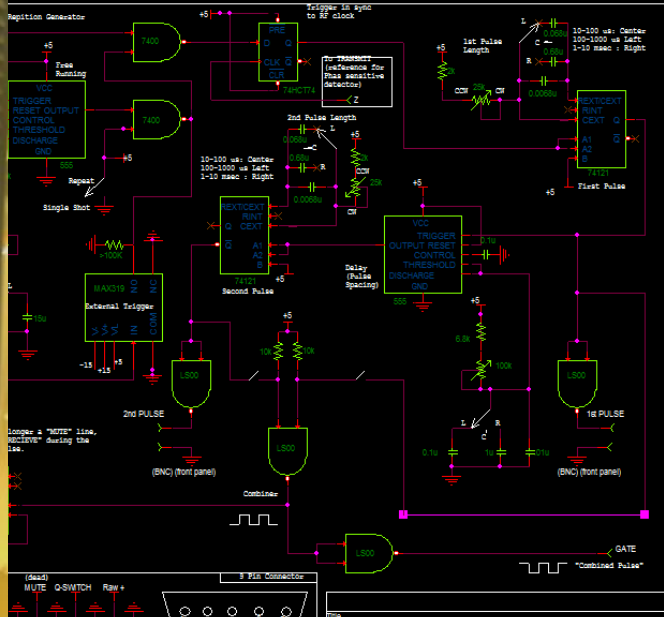
- Solutions? For binary cross sections

$$\sigma(T) = \frac{1}{(kT)^2} \int_0^\infty dE \sigma(E) E e^{-E/kT}.$$

$$\begin{aligned} \sigma(E) = & \frac{8\pi\mu^2}{3\hbar^4} \int_0^\infty b^3 db \\ & \times \left| \int_{r_0}^\infty dR \frac{c_K(R)}{\sqrt{1 - b^2/R^2 - V(R)/E}} \right|^2 \end{aligned}$$

Low-field NMR

- Result of first project with group

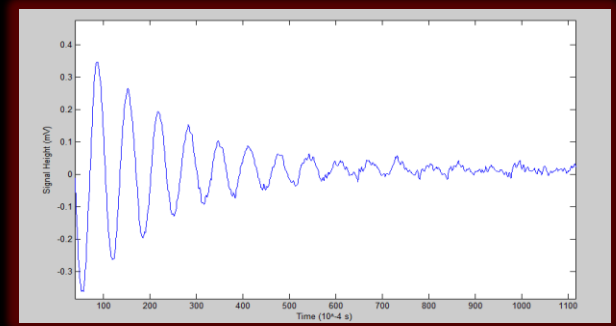


```

function [datapoints,phasedft,freq] = compute(file,upordown , addnum , t_FID , apod_num, MBEFID,cutoffbegFID , cutoffFID)
clearvars -except filename addnum t_FID apod_num MBEFID cutoffFID upordown cutoffbegFID;
[Pro_Data,~,freq] = prodata(filename, addnum , t_FID , apod_num);
[fft_data] = buildspecs(Pro_Data);
siz = size(fft_data,2);
tic;
phasedft = zeros(size(fft_data,1),siz-cutoffFID-cutoffbegFID);
datapoints = zeros(1,siz-cutoffFID-cutoffbegFID);
for n = 1:(siz-cutoffFID-cutoffbegFID)
    %tic
    phasedft(:,n) = autophase(fft_data(:,n),cutoffbegFID,cutoffFID);
    datapoints(n) = int(fft(phasedft(:,n)));
    %str = 'Autophasing and integration took ',num2str(toc),' seconds for iteration ', num2str(n) ,' out of ', num2str(siz) ;
    %disp(str)
end
str = ['Autophasing and integration took ',num2str(toc),' seconds' ]; disp(str);
datapoints = datapoints/max(datapoints);
t_1 = (1:length(datapoints))*MBEFID - MBEFID;
[~,t_1] = fitted(datapoints,t_1,upordown);
% hold on
% if
%plot(t_1/60, datapoints , '.', t_1, E, '-');
% hold off
end

```

Analysis



Free Induction Decays (FID)

- Collect 20-120 FID's for a given T₁ measurement
- Baseline correct and zerofill FID's
- FFT FID's
- Phase correct FFT'S
- Integrate phase corrected FFT's
- 2-parameter fit these to exponential decay
- T₁ (or relaxation rate) comes from this

Preliminary Results

- Coatings from Geoff Schrank at PNNL

Cell 151A
8-member
fluorocarbon,
Pure xenon

| Temp (deg C) | T ₁ (hrs) | T ₁ _wall (hrs) |
|--------------|----------------------|----------------------------|
| 150 | 3.9 ± .7 | 9.7 ± 2.6 |
| 150 | 1.2 ± .5 | 1.5 ± .7 |
| 150 | 1.8 ± .1 | 2.4 ± .2 |

Cell 151B
18-member
hydrocarbon,
Pure xenon

| Temp (deg C) | T ₁ (hrs) | T ₁ _wall (hrs) |
|--------------|----------------------|----------------------------|
| 150 | 4.8 ± .7 | 15 ± 3 |
| 150 | 4.1 ± .4 | 9.6 ± 1.3 |
| 25 | 2.7 ± .4 | 8.6 ± 1.5 |
| 26 | 2.5 ± .4 | 6.9 ± 1.3 |
| 175 | 4.7 ± .1 | 12 ± 1.2 |

Preliminary Results

- In-house coatings

Cell 151C
8-member
hydrocarbon,
pure xenon

| Temp (deg C) | T ₁ (hrs) | T ₁ _wall (hrs) |
|--------------|----------------------|----------------------------|
| 150 | 3.3 ± .2 | 6.4 ± .7 |
| 150 | 3.2 ± .2 | 6.2 ± .7 |
| 150 | 2 ± 1 | 5.3 ± 2.7 |
| 150 | 2.7 ± 2 | 4 ± 3 |
| 150 | 3.2 ± .1 | 6.2 ± .6 |
| Cell | Broken | |

Preliminary Results

- Re-coated 151A & 151B with 18-member hydrocarbon in-house (OCD runs)

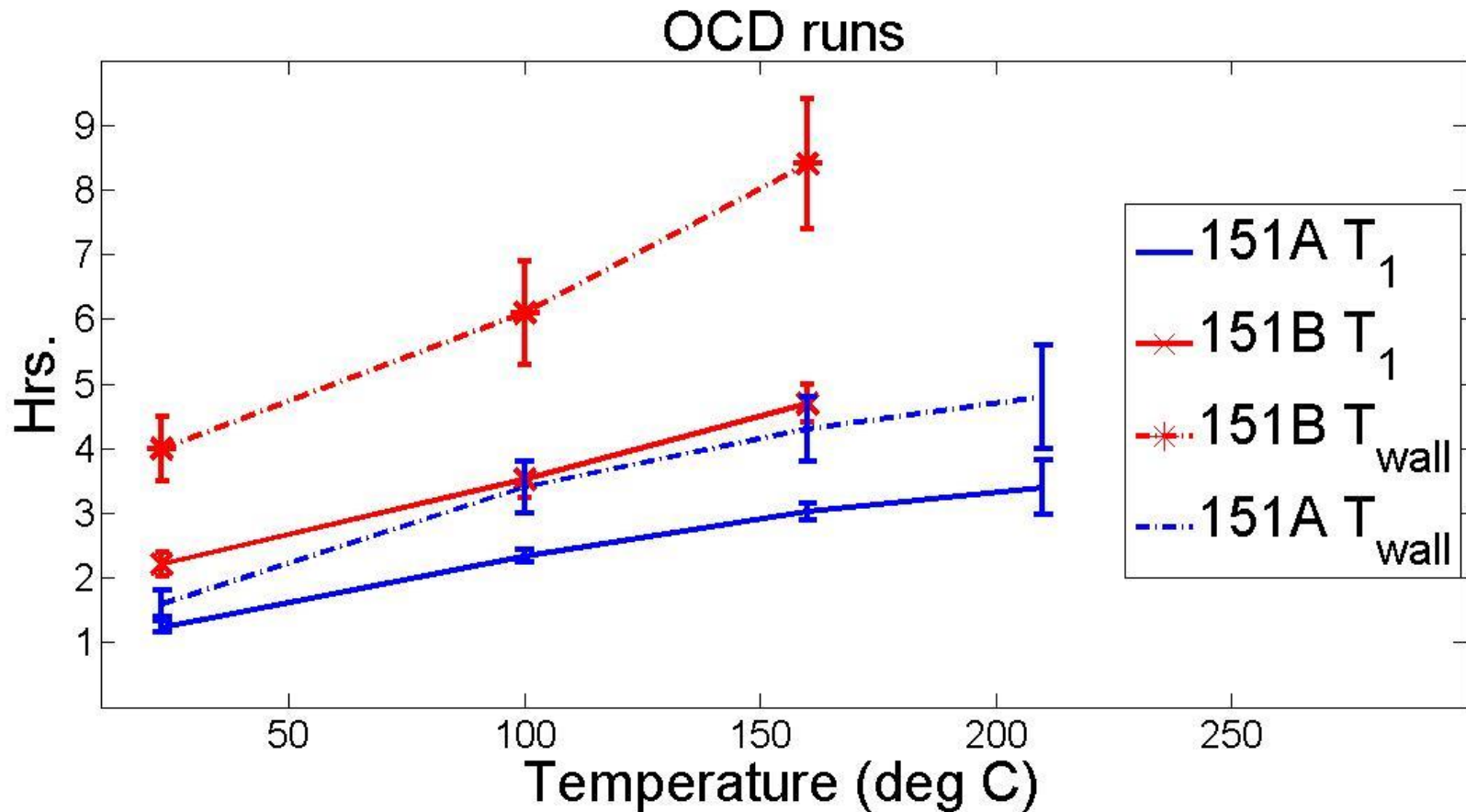
T₁

| Cell | Coating | 23°C | 100°C | 160°C | 210°C |
|------|---------|-----------|-----------|-----------|-----------|
| 151A | OCD(18) | 1.24(.09) | 2.33(.10) | 3.02(.12) | 3.39(.42) |
| 151B | OCD(18) | 2.21(.18) | 3.52(.28) | 4.69(.29) | |
| BL1 | OCD(18) | .93(.05) | | | |

T_{wall}

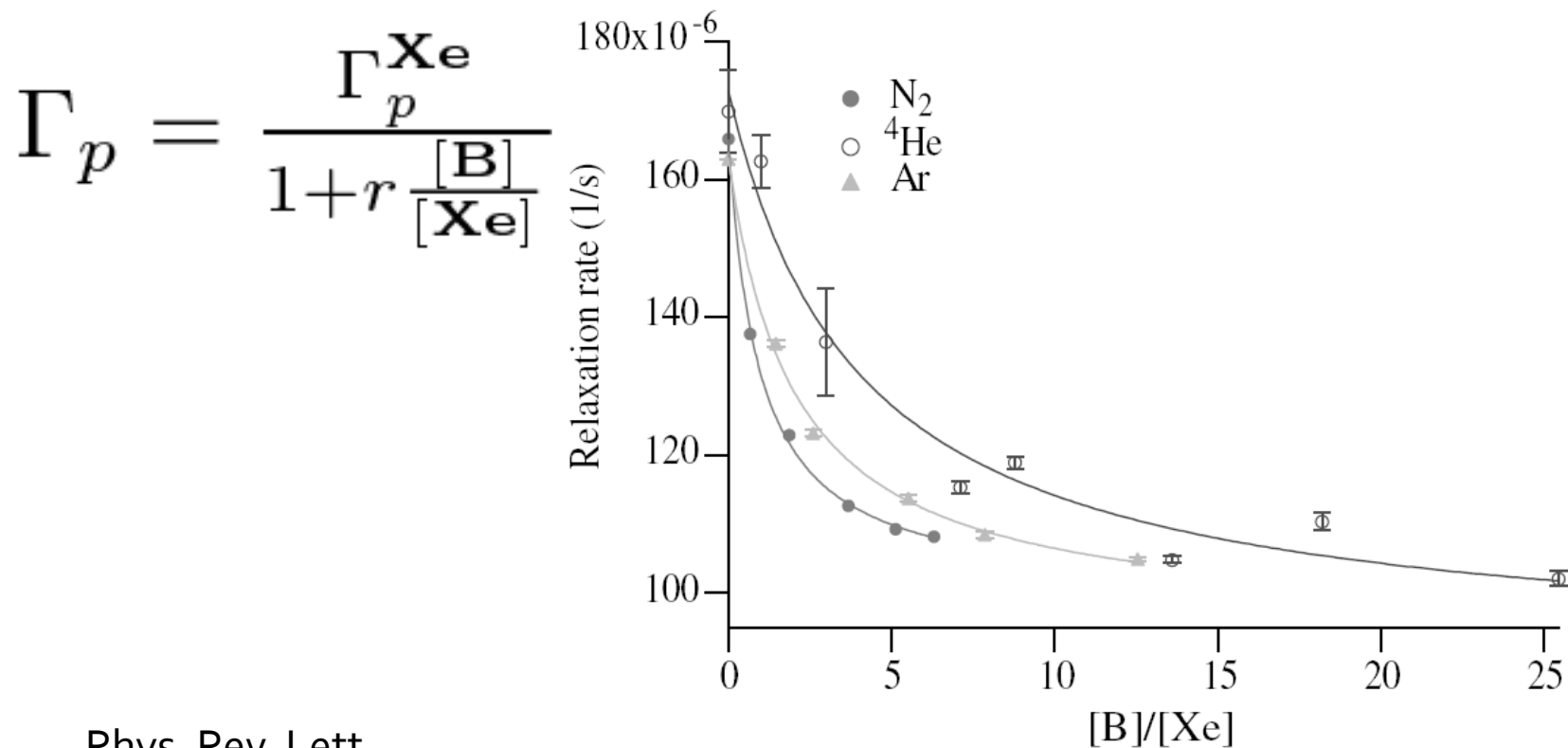
| Cell | Coating | 23°C | 100°C | 160°C | 210°C |
|------|---------|---------|---------|----------|---------|
| 151A | OCD(18) | 1.6(.2) | 3.4(.4) | 4.3(.5) | 4.8(.8) |
| 151B | OCD(18) | 4.0(.5) | 6.1(.8) | 8.4(1.0) | |
| BL1 | OCD(18) | | | | |

Preliminary Results



Introducing Nitrogen

- Buffer gas helps yet again



Phys. Rev. Lett.
Chann et. al. 2002

FIG. 1. Xe spin-relaxation rate as a function of composition, for various buffer gases, at a fixed Xe density of 0.15 amagat.

Characterizing Coatings

- Find T_1 in with pure xenon in cell at low-field
- Introduce high pressure of nitrogen into cell with pure xenon
- Sweep temperature range of coating
- Back out the wall relaxation
- Push temperature limit of coating
- Purposefully degrade coatings to test durability (haven't done... on purpose)

Current and Future Projects

- Surprising T_1 phenomenon seen in solid xenon, i.e. see a lengthening in T_1 , seemingly dependent on passing through the liquid phase

| Temp (K) | Snow T_1 (min) | Ice T_1 (min) |
|----------|------------------|-----------------|
| 77 | 142 ± 6 | 173 ± 6 |
| 122 | 45 ± 15 | |
| 132 | 36 ± 8 | 61 ± 9 |
| 147 | 35 ± 3 | 50 ± 3 |

Current and Future Projects

- Is it structural? FCC-HCP combinations or many broken lattices as some groups are claiming?

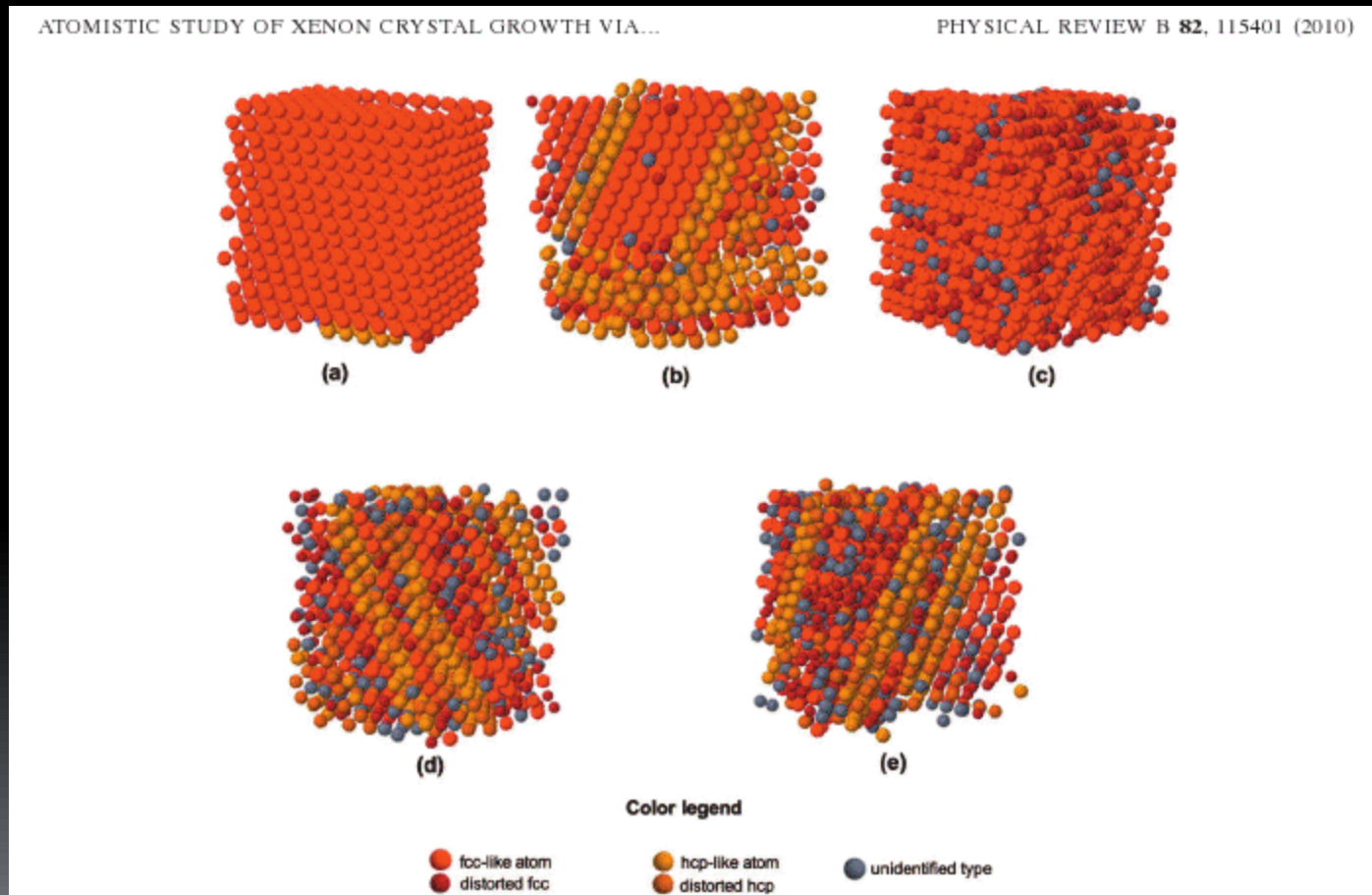


FIG. 6. (Color) Bond angle analysis performed on structures obtained as a result of tempering hot samples as obtained after a simulated deposition with a rate of 2×10^{11} atoms/s. Only 12-fold-coordinated Xe atoms are shown. Tempering temperatures and simulation times are as follows: (a) 10 K, 64 ns; (b) 25 K, 72 ns; (c) 50 K, 110 ns; (d) 75 K, 140 ns; and (e) 100 K, 56 ns.

Conclusion

- Hyperpolarized noble gases make unique systems for NMR study.
- Polarizer is a unique tool that gives us a way to isolate hyperpolarized xenon...
- But...
- Need to develop a way to store the gas for extended periods of time.
- Projects in Spintronics, Medical Physics, and Dilute Xenon Spins coming soon!

