Spatial Polarization Profile in an Optically Pumped Alkali Vapor

Ben Andrew Olsen, Brian Patton*, Yuan-Yu Jau, Will Happer

Department of Physics, Princeton University
*Now at the Department of Physics, UC Berkeley

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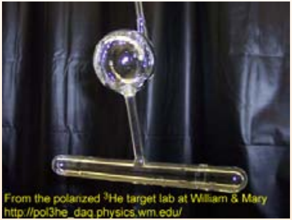
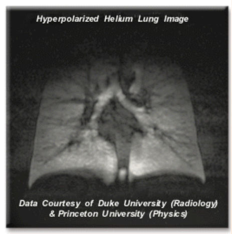
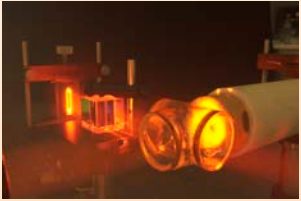
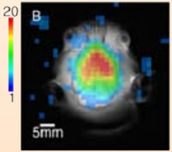
University of Virginia

Applications of hyperpolarized substances









Rat brain image borrowed from Swanson *et al.*, Magnetic Resonance in Medicine 38(5), 695

^{133}Cs NMR enhancement in solid CsH

- $\sim 8\times$ enhancement of ^{133}Cs NMR signal in solid CsH [Ishikawa, *et al.*, PRL **98**, 183004 (2007)]
- Modest enhancement of ^{133}Cs NMR signal in solid CsCl, ^{1}H NMR signal in CsH [DAMOP (2008)]

- Enhancement is not repeatable—identically prepared cells give wildly different results
- Both electronic and nuclear spin currents seem to contribute
- We want to understand interactions and dynamics of the optically pumped alkali vapor
  - Spatial dependence
  - Pump frequency dependence
  - Pump power, magnetic field, buffer gas pressure/species, time...

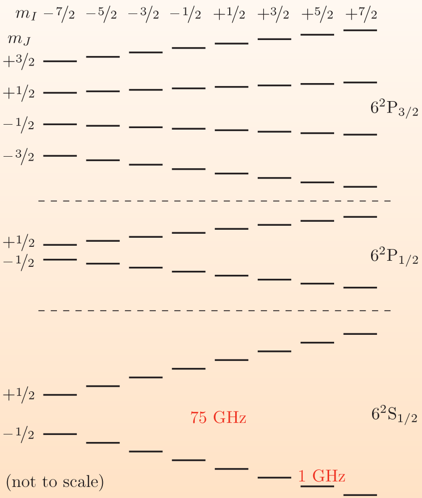
Alkali metal atom energy levels

- A single-electron atom with nuclear spin $\mathbf{I}$ and electronic spin $\mathbf{J} = \mathbf{L} + \mathbf{S}$ in an external magnetic field $\mathbf{B}$ is described by the Hamiltonian:

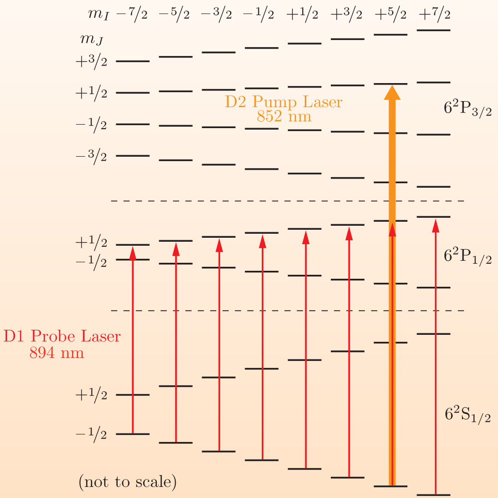
$$\mathcal{H}_{L,J} = E_{L,J} + g_{L,J}\mu_B \mathbf{J} \cdot \mathbf{B} + \mathcal{A}_{L,J} \mathbf{I} \cdot \mathbf{J} + g_I \mu_B \mathbf{I} \cdot \mathbf{B} + \mathcal{H}_{\mathrm{QP}}$$

- $E_{L,J}, g_{L,J}, \mathcal{A}_{L,J}$ are the term energy, g-factor, and hyperfine coupling constant for a given level
- The nuclear Zeeman and quadrupole terms are small, so let's ignore them

Energy levels in ^{133}Cs at 2.7 T



Optical pumping and spectroscopy in ^{133}Cs at 2.7 T



Absorption due to alkali vapor

On resonance, the relative population $\mathcal{P}$ of a hyperfine level is related to the transmitted intensity in a cell of length H by:

$$\begin{aligned}\mathcal{P} &= -\ln\left(\frac{I}{I_0}\right)\left(\frac{1}{\sigma_{\text{abs}}\,[\text{Cs}]\,H}\right)\\ &= \alpha\ln I + \beta\,.\end{aligned}$$

We can use this simple relation to infer $\mathcal{P}$ for each ground state sublevel with $m_J = -1/2$ from an absorption spectrum.

Simplified spin diffusion model

If we treat the pumped sublevel as independent from all the other sublevels, the relative polarization $\mathcal{P}$ obeys the diffusion equation:

$$\frac{\partial \mathcal{P}}{\partial t} + \mathcal{D}\nabla^2\mathcal{P} = -\Gamma_{\mathrm{op}}\mathcal{P}\,,$$

Where Γ_{op} is the pumping rate and $\mathcal{D}$ is the diffusion coefficient for Cs in N_2 gas. At steady state, with $f = 1 - \mathcal{P}$, we have

$$\mathcal{D}\nabla^2 f + \Gamma_{\mathrm{op}}(1 - f) = 0\,.$$

In an infinite cylinder with $f = 0$ at the walls $(r = R)$, we have

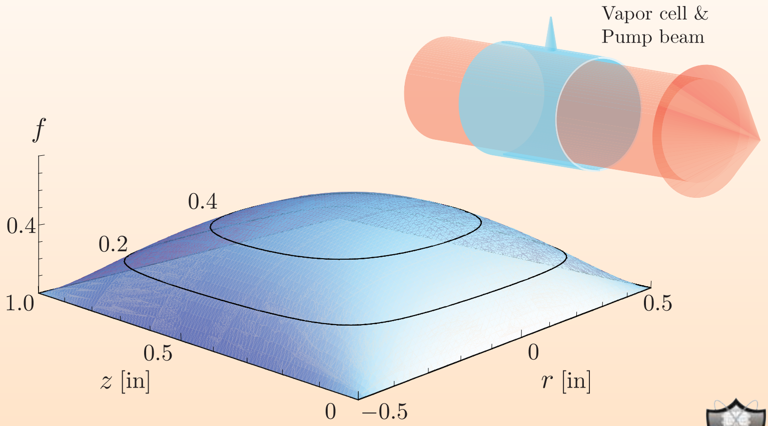
$$\mathcal{P}(r) = 1 - f(r) = \frac{I_0(r\sqrt{\Gamma_{\mathrm{op}}/\mathcal{D}})}{I_0(R\sqrt{\Gamma_{\mathrm{op}}/\mathcal{D}})}\,,$$

where I_0 is a modified Bessel function.

Numerical simulation of spin diffusion

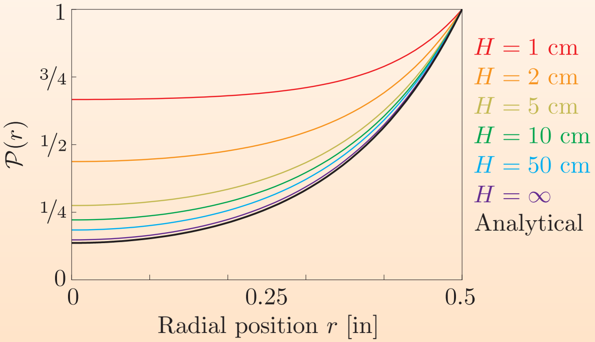
- Initialize a grid of 50×50 points, representing the region $(0 < r < R, \phi = 0, 0 < z < H)$, with $h_r = R/50, h_z = H/50$
- Assign each point a new value based on the old values of its neighbors:
  $f_{i,j} = \frac{h_r^2(f_{i,j-1}+f_{i,j+1})+h_z^2(1+R/2ih_r)f_{i+1,j}+h_z^2(1-R/2ih_r)f_{i-1,j}+h_r^2\ h_z^2\ \Gamma_{\mathrm{op}}/\mathcal{D}}{2h_r^2+2h_z^2+h_r^2\ h_z^2\ \Gamma_{\mathrm{op}}/\mathcal{D}}$
- Enforce boundary conditions: $f = 0$ for $z = 0, z = H, r = R$, and $\partial f/\partial r = 0$ for $r = 0$
- Keep iterating until the values on the grid don't change much

Simulated polarization along a diameter



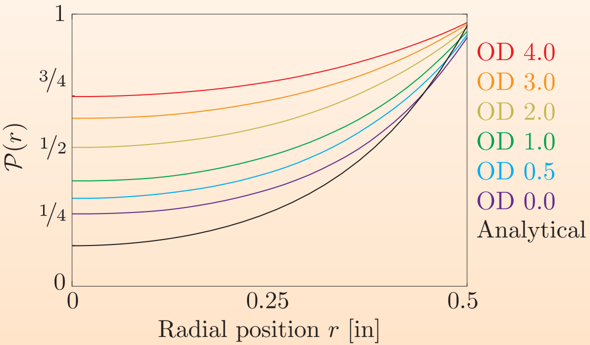
Simulated polarization profile (averaged along the vertical)

$$\Gamma_{\text{op}} = 50\ \text{s}^{-1},\ \text{OD} = 0,\ R = 0.5\ \text{in}$$



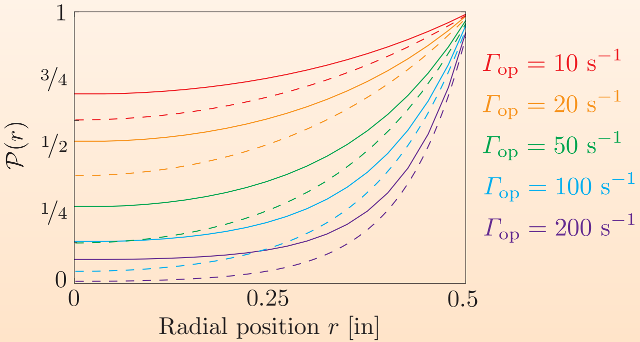
Simulated polarization profile (averaged along the vertical)

$$\Gamma_{\mathrm{op}} = 50\ \mathrm{s}^{-1}, H = 2\ \mathrm{in}, R = 0.5\ \mathrm{in}$$

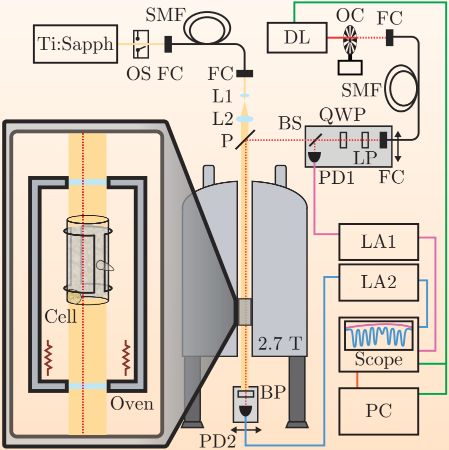


Simulated polarization profile (averaged along the vertical)

$\mathrm{OD} = 1, H = 2$ in, $R = 0.5$ in (experimental conditions)



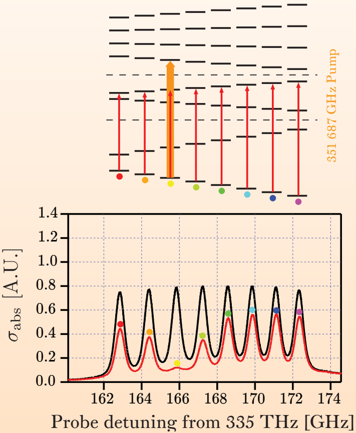
Experimental setup

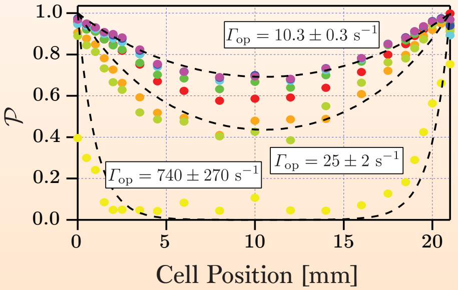


Experimental procedure

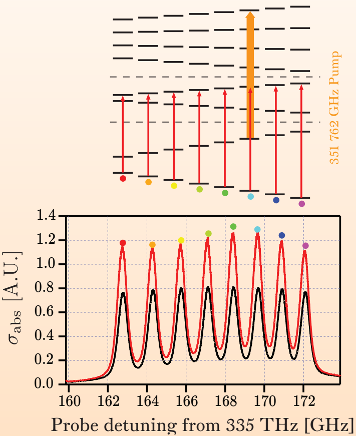
- Scan the probe laser both with the pumping light off and with the pumping light on
  - The pumping light (D2) is tuned to depopulate a single sublevel
  - The probe (D1) measures the population of each sublevel in the $m_S = -1/2$ multiplet.
- Acquire a spectrum pair at many positions in the cell, moving in multiples of 0.5 mm, starting at one cell wall and ending at the other
  - Mechanical translation stage moves beam horizontally
  - Second translation stage moves detection optics

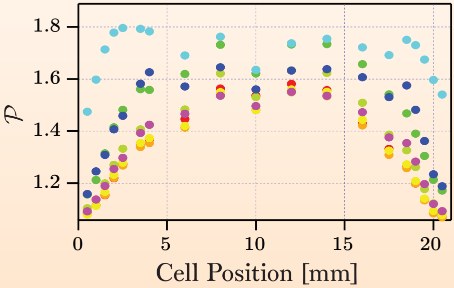
Polarization profile for an allowed transition (10 Torr N_2, 0.7 mW)



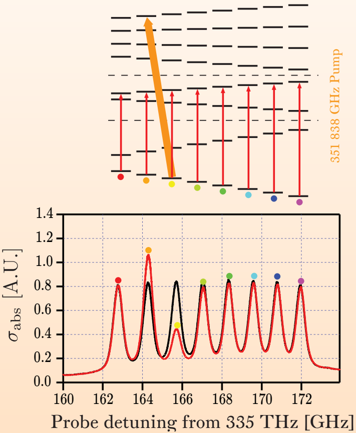


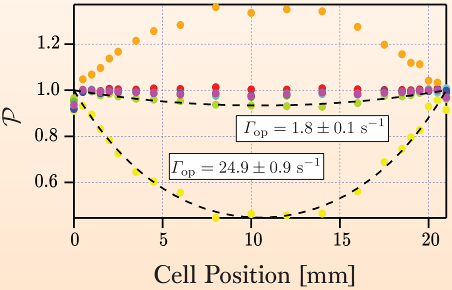
Polarization profile for the conjugate transition



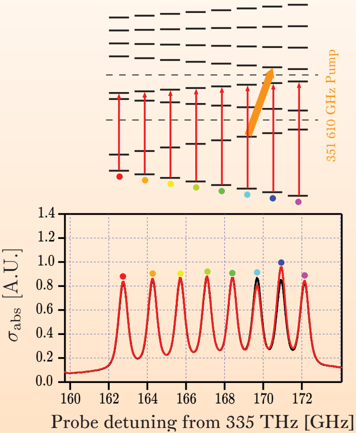


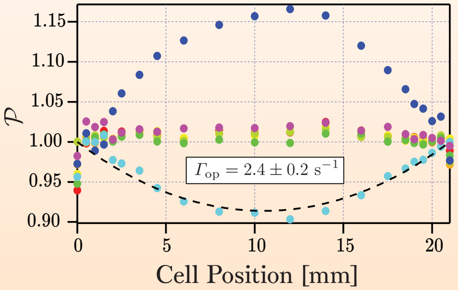
Polarization profile for a forbidden transition



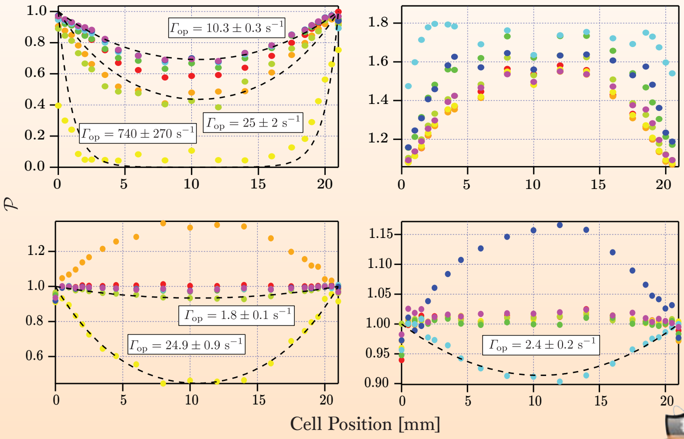


Polarization profile for the conjugate transition

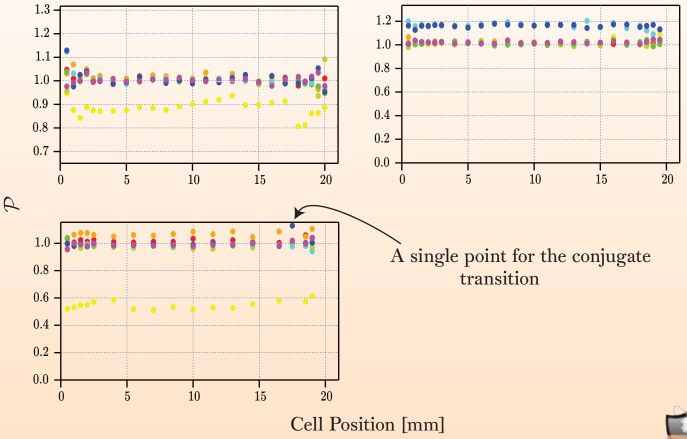




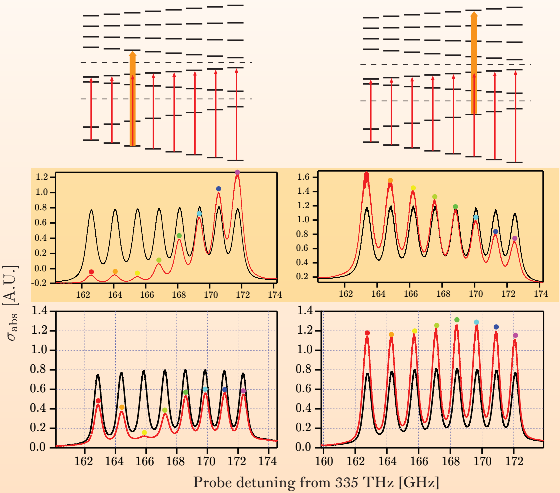
Polarization profiles for all 4 transitions (10 Torr N_2, 0.7 mW)



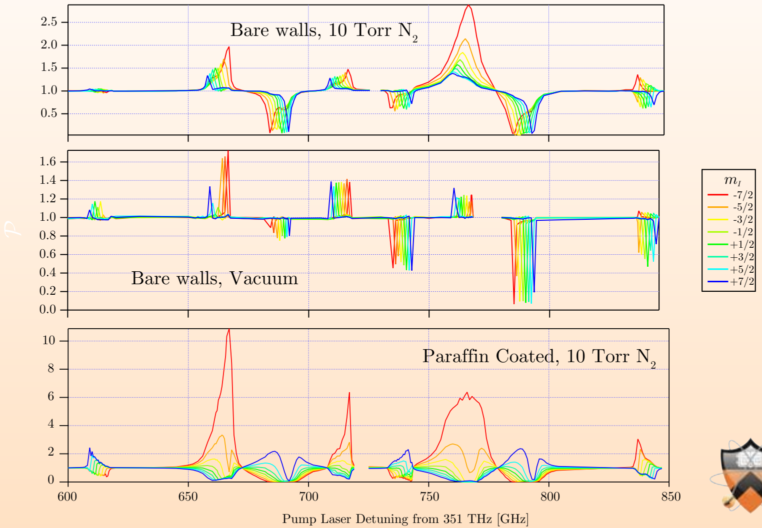
Polarization profiles for all 4 transitions (Vacuum, 0.7 mW)



Paraffin-coated cell (10 Torr N_2, 0.7 mW)



Populations as functions of pump frequency



Conclusion

- Simulated spin diffusion in finite vapor cells
- Measured cesium vapor absorption spectra at 2.7 T
    - Individual sublevel populations obey a diffusion equation with complete depolarization at the cell walls (except with no buffer gas or with paraffin coating)
    - Pumping rates for detuned transitions may tell us about spin exchange in the vapor
- Cells with no buffer gas and with paraffin coatings exhibit strange behavior

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Vapor Cells

Some more Data