Basics of rubidium-xenon spin-exchange optical pumping

Magnetising xenon gas

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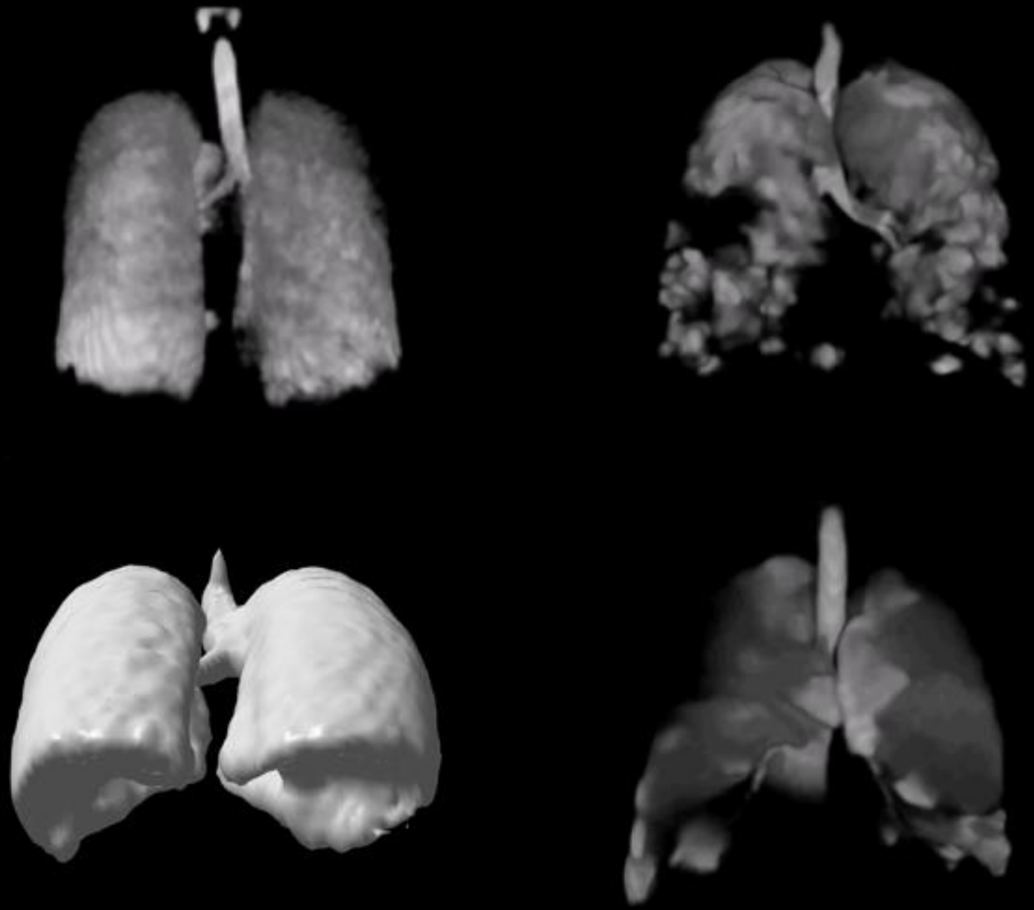
University of Sheffield

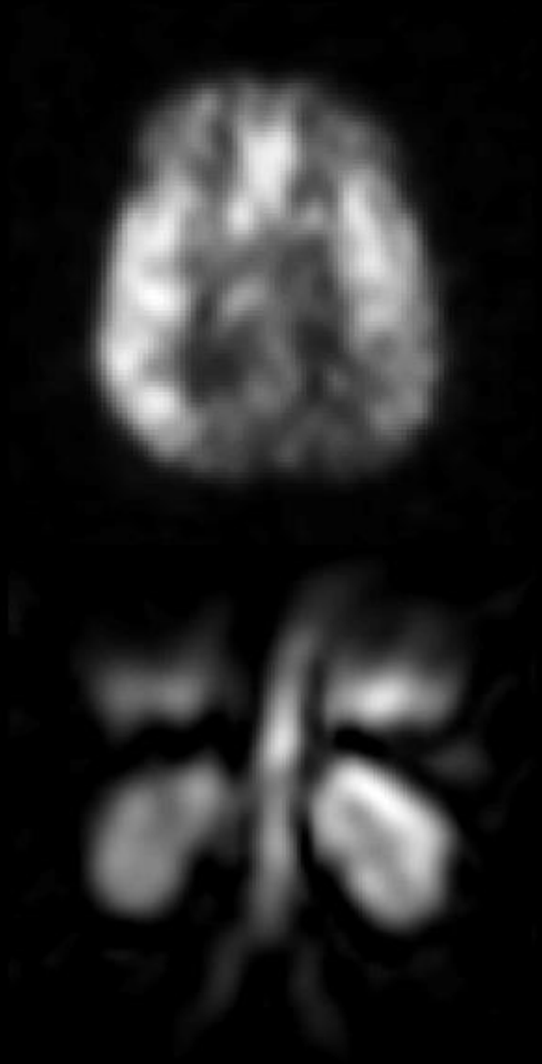
POLARIS
Polarised lung and respiratory imaging Sheffield
POLARised Imaging Systems

Hyperpolarised ^{129}Xe MRI

Healthy COPD



Gas-phase ^{129}Xe



Human brain

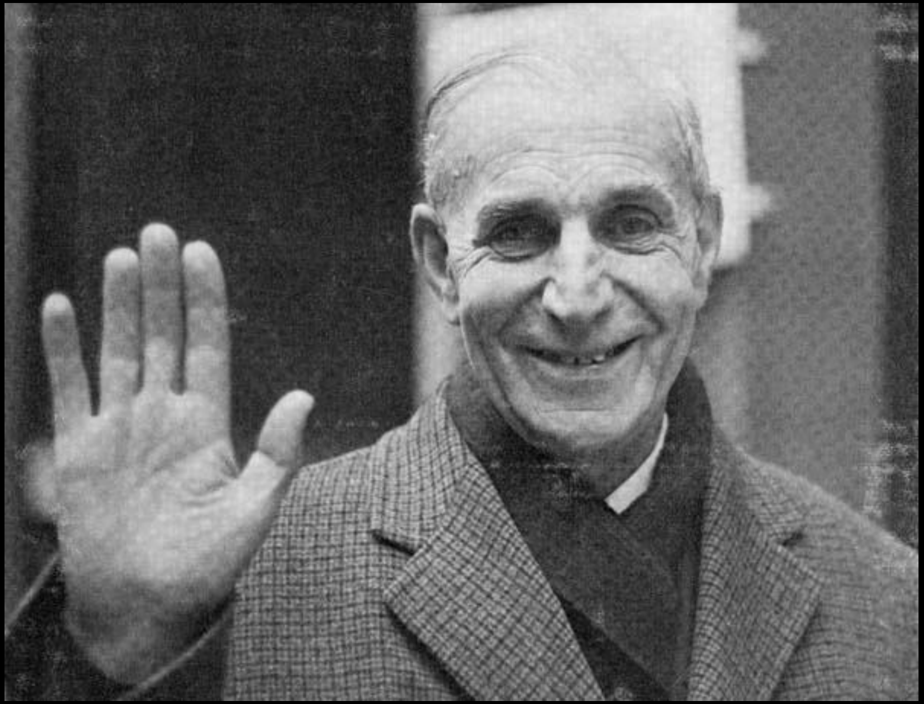
Rao et al, Radiology, 286, 2018

Human kidneys

Chacon-Caldera et al., MRM(83) 2019

Dissolved-phase ^{129}Xe

Optical pumping



Alfred Kastler

Awarded 1966 Nobel prize in physics for his work

A. Kastler. "Some suggestions concerning the production and detection by optical means of inequalities in the populations of levels of spatial quantization in atoms. Application to the Stern and Gerlach and magnetic resonance experiments. *J. Phys. Radium*, **11**, 255. (1950)

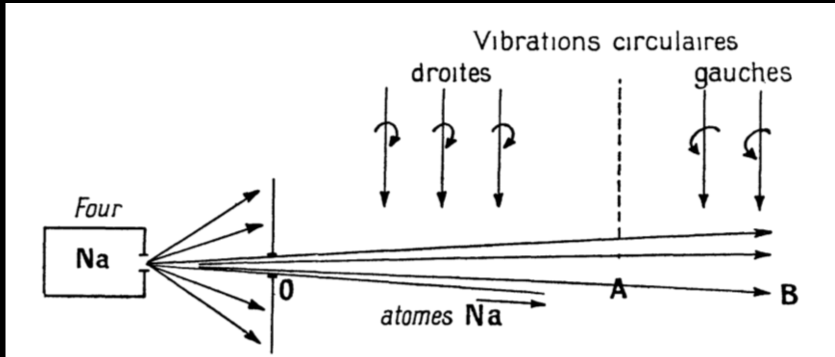


Fig. 9.

"The use of circularly polarised light creates an asymmetry of population between negative m levels and positive m levels, the direction of this asymmetry being able to be reversed by reversing the direction of circular polarisation of the incident light"

Optical pumping with alkali metals

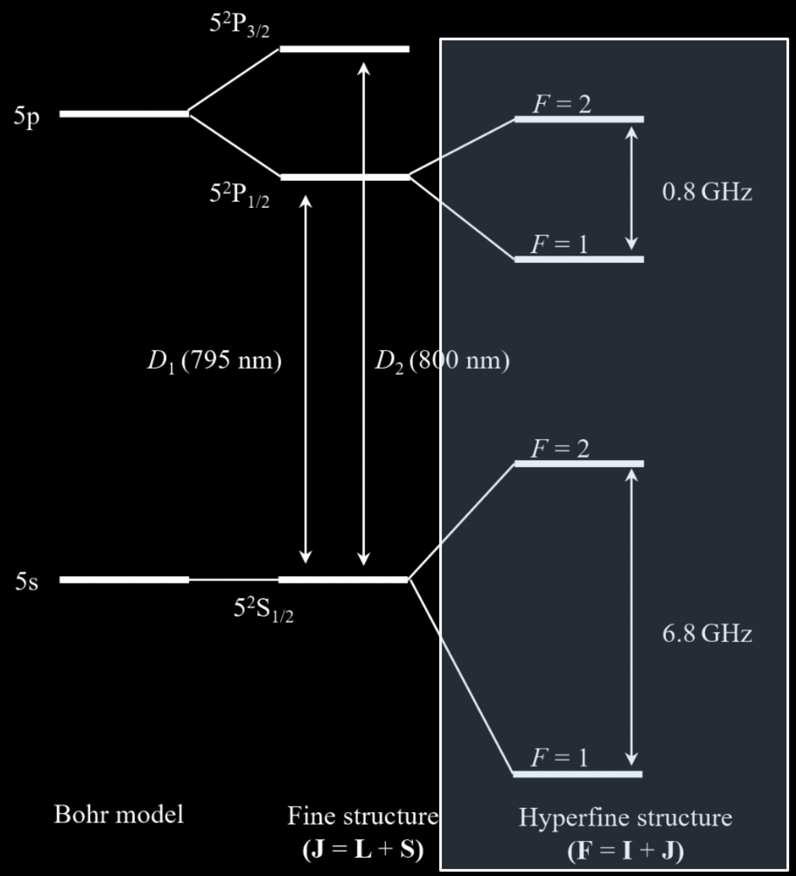
- λ in visible and near-IR (590-865 nm) – compatible with inexpensive and powerful laser sources
- High volatility – highly dense vapours at relatively low temperatures [MP = 27°C (Fr) to 181°C (Li)]
- Single valence electron in *s* subshell – facilities comparison between theory and experiment

	Li	Na	K	Rb	Cs	Fr
Z	3	11	19	37	55	87
Configuration	[He]2s	[Ne]3s	[Ar]4s	[Kr]5s	[Xe]6s	[Rn]7s
IP (eV)	5.3917	5.1391	4.3407	4.1771	3.8939	4.073
$\lambda(S\text{-}P_{1/2})$ (Å)	6709.77	5897.56	7701.08	7949.78	8945.93	8171.66
$\lambda(S\text{-}P_{3/2})$ (Å)	6709.62	5891.58	7667.01	7802.41	8523.47	7181.85
ΔE_{FS} (cm^{-1})	0.34	17.20	57.71	237.60	554.04	1687
$\tau(nP_{1/2})$ (ns)	27.10 [8]	16.28 [9]	26.69 [10]	27.75 [11]	34.88 [12]	29.45 [13]
$f_{1/2}$	0.249	0.320	0.333	0.341	0.344	0.339
$\tau(nP_{3/2})$ (ns)	27.10 [8]	16.23 [9]	26.34 [10]	26.25 [11]	30.462 [12]	21.0 [14]
$f_{3/2}$	0.497	0.641	0.669	0.695	0.715	0.734
μ_{ns}	$0.399 - 0.061E$	$1.348 - 0.132E$	$2.180 - 0.314E$	$3.130 - 0.419E$	$4.048 - 0.573E$	$5.07 - 0.556E$
μ_{np}	$0.047 + 0.050E$	$0.854 - 0.258E$	$1.711 - 0.542E$	$2.645 - 0.681E$	$3.568 - 0.889E$	
μ_{nd}	$0.002 + 0.009E$	$0.015 + 0.081E$	$0.278 + 2.137E$	$1.353 + 1.760E$	$2.476 + 0.372E$	$3.41 - 0.095E$

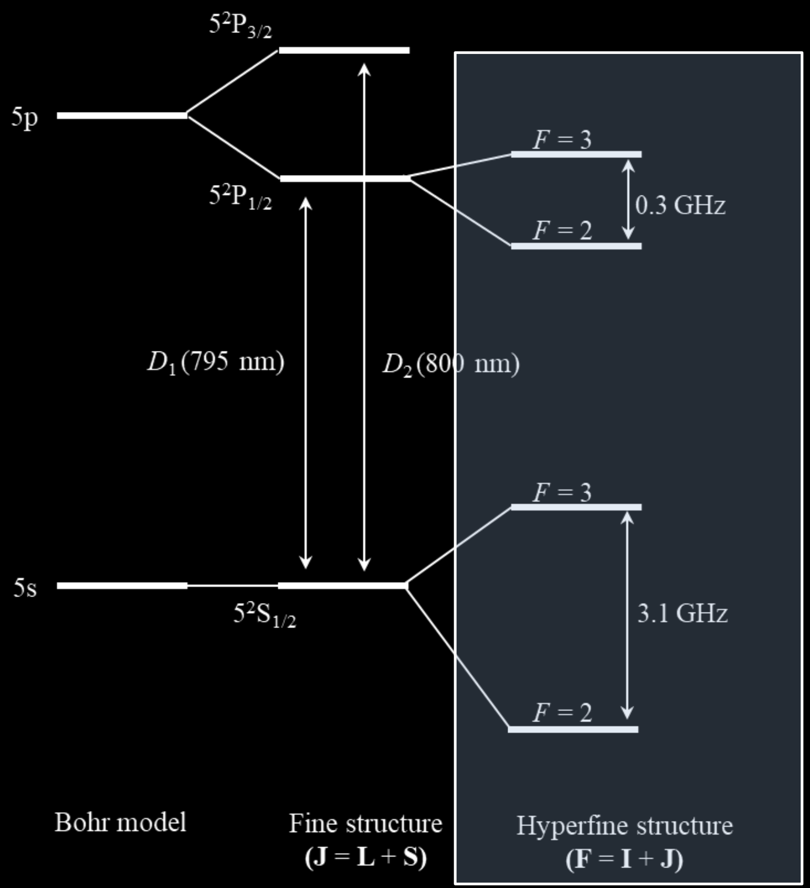
Energy states of rubidium

- Abundance: ^{87}Rb (27.8%), ^{85}Rb (82.2%)
- Nuclear spin: $I_{87} = 3/2, I_{85} = 5/2$

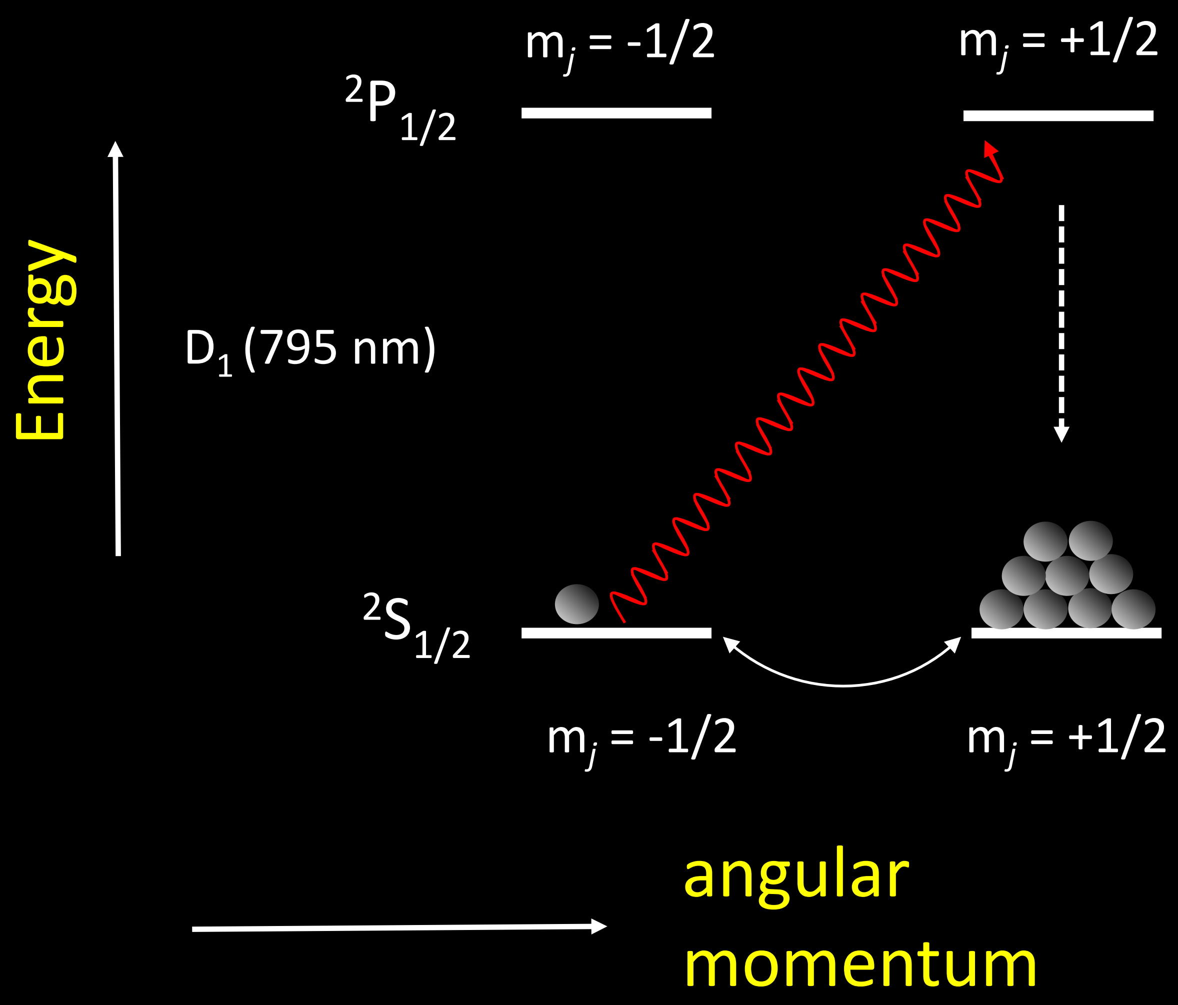
^{87}Rb



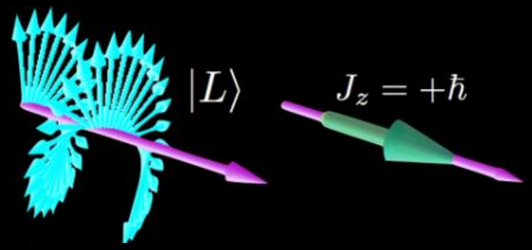
^{85}Rb



Optical pumping with rubidium



Left-circularly polarised light parallel to B_z

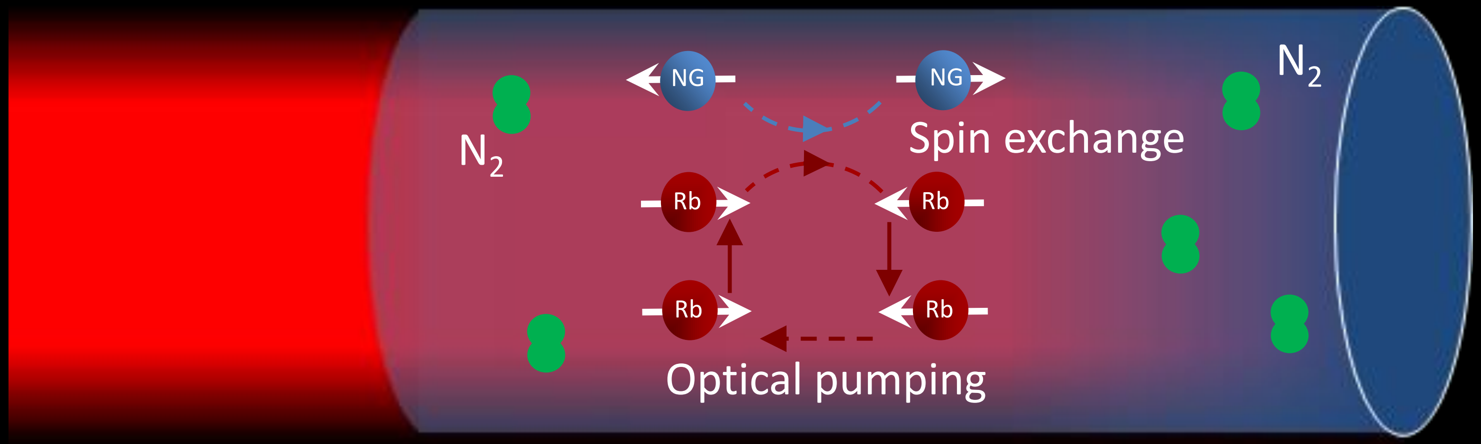


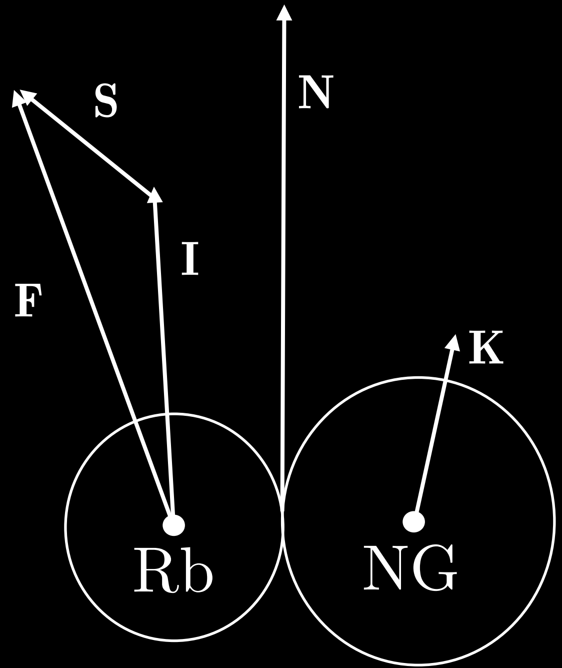
Selection rule:

$\Delta m_j = +1$

Mixing polarised rubidium vapour with noble gases: SEOP

Circularly polarised light



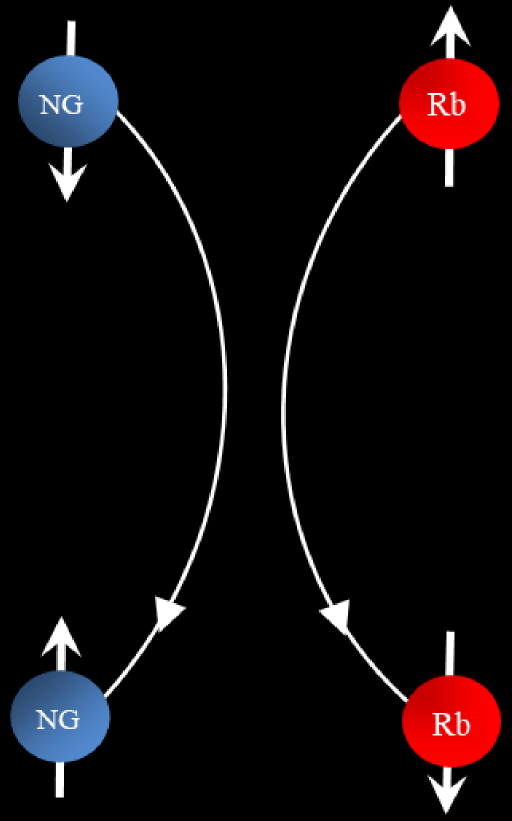


$$H = A\mathbf{I}\cdot\mathbf{S} + \gamma\mathbf{N}\cdot\mathbf{S} + \alpha\mathbf{K}\cdot\mathbf{S} + \cdots$$

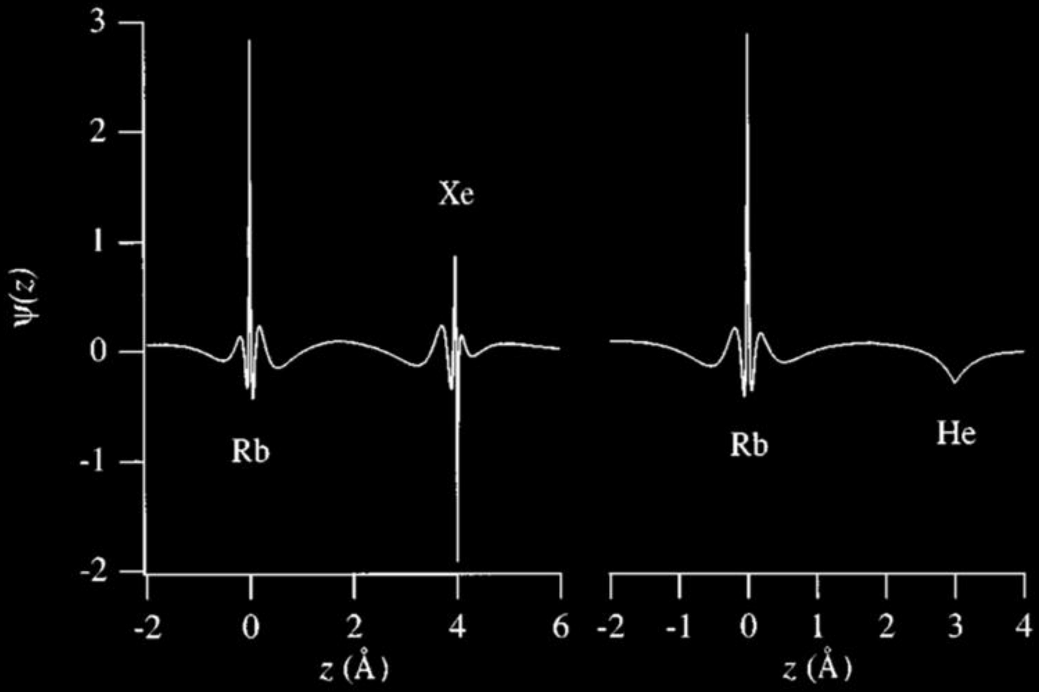
Spin exchange

Bouchiat et al., PRL(5), 1960; Grover PRL(40) 1978

Spin-exchange optical pumping with noble gases



$\langle K \cdot S$



Walker&Happer RMP(69), 1997

Binary spin-exchange rate:

$$\gamma_{se} = [\text{Rb}]\gamma' \approx 10^{14} \times \begin{cases} 6.8 \times 10^{-20}\text{cm}^3/\text{s} & \text{He} \\ 2.2 \times 10^{-16}\text{cm}^3/\text{s} & \text{Xe} \end{cases}$$

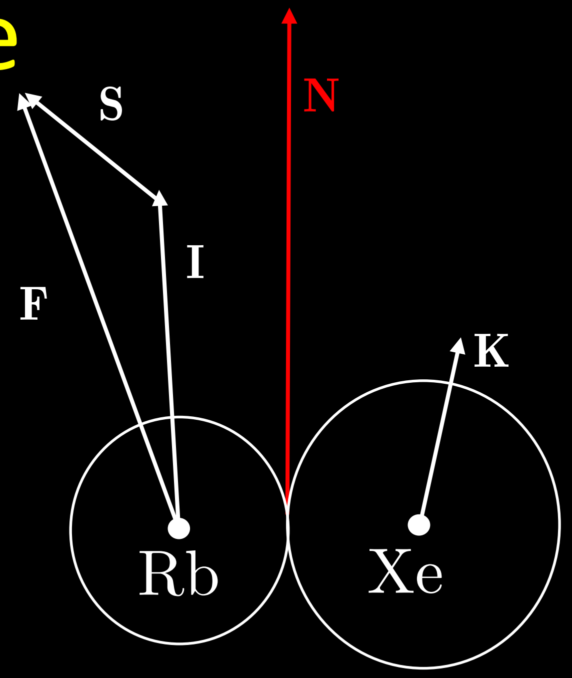
$$\approx \begin{cases} (40\ \text{hr})^{-1} & \text{He} \\ (50\ \text{sec})^{-1} & \text{Xe} \end{cases}$$

^{129}Xe readily destroys Rb electronic polarisation

$$\Gamma_{\mathrm{Rb}}(1\ \mathrm{amagat}) = \begin{cases} 2.4 \times 10^5/\mathrm{s} & \text{Xe} \\ 45/\mathrm{s} & \text{He} \end{cases}$$

$$H = A\mathbf{I}\cdot\mathbf{S} + \gamma\mathbf{N}\cdot\mathbf{S} + \alpha\mathbf{K}\cdot\mathbf{S} + \cdots$$

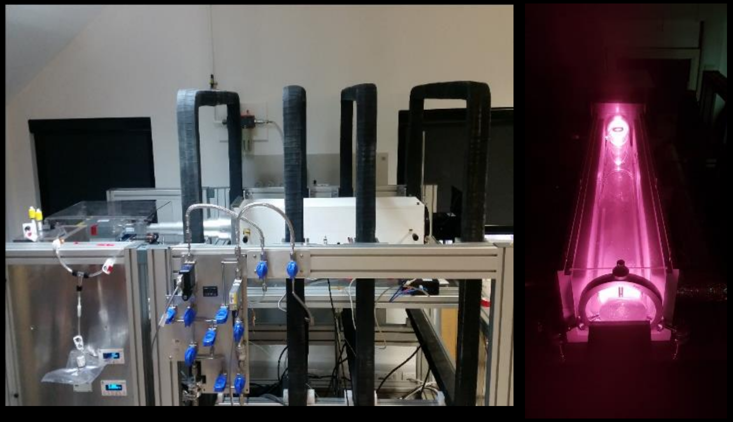
Spin-rotation interaction at xenon core



Challenge: need to use lean mixtures of Xe or low Rb vapour densities

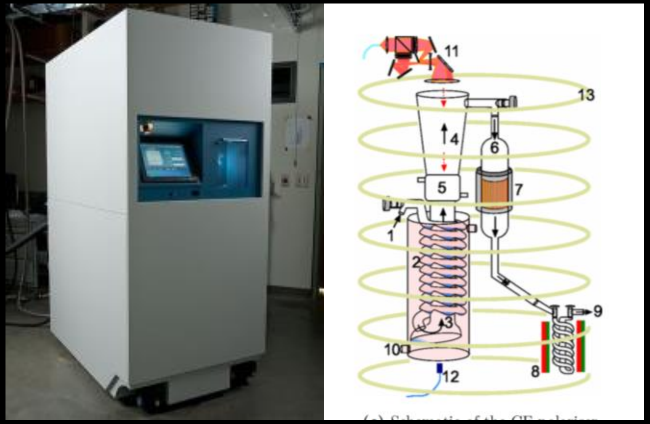
Two methods to polarise ^{129}Xe with spin-exchange optical pumping

Continuous-flow: ^{129}Xe polarised with cryogenic Xe accumulation



Norquay et al., PRL(121), 2018

Polarean 9820



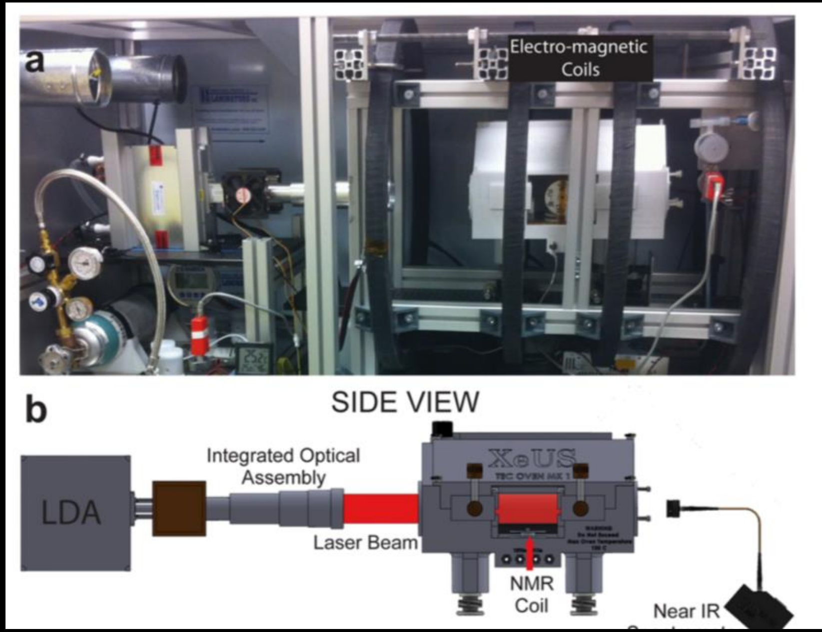
Xemed, XeBox-E10, Ruset et al., PRL(96), 2006

Other continuous-flow polarisers

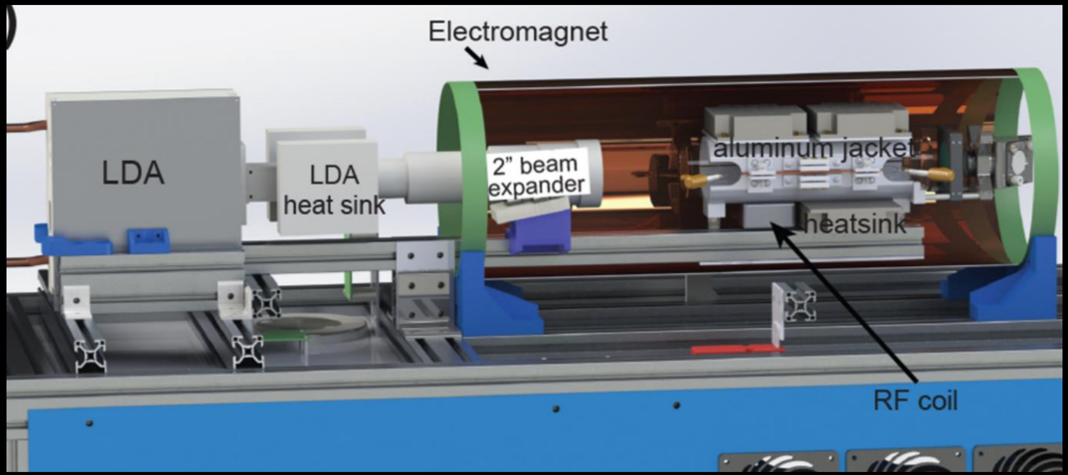
Driehuys et al, APL(69), 1996; **Haake et al.**, JACS(119), 1997; **Rosen et al.**, RSI(70), 1999; **Zook et al.**, JMR(159), 2002; **Mortuza et al.**, JCP(118), 2003; **Knagge et al.**, CPL(397), 2004; **Schrank et al.**, PRA(80), 2009; **Norquay et al.**, JAP(113), 2013. Korchak et al., AMR(44), 2013.

Two methods to polarise ^{129}Xe with spin-exchange optical pumping

Stopped-flow: ^{129}Xe polarised without cryogenic Xe accumulation



Nikolaou et al., JPC(118), 2014.



Birchall et al., AC(92), 2020.

Other stopped-flow polarisers

Birchall et al., Molecules(27), 2022; **Raftery et al.**, PRL(66), 1991; **Whiting et al.**, JMR(208), 2011; **Six et al.**, PloS one(7), 2012; **Hughes-Riley et al.**,JMR(237), 2013; **Nikolaou et al.**, AC(86), 2014; **Birchall et al.**, JMR(315), 2020.

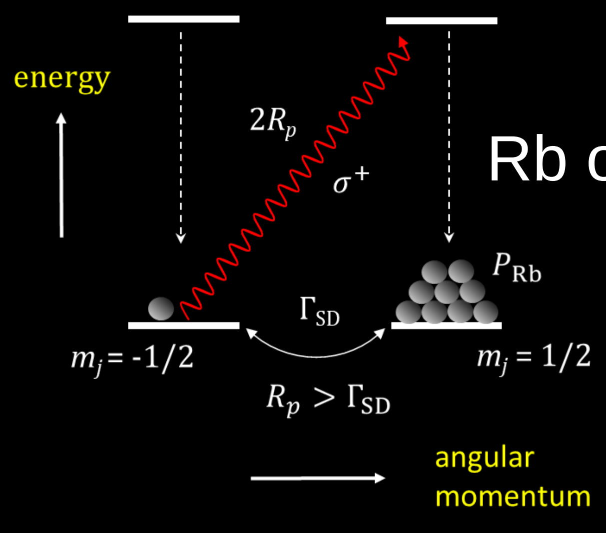
Stopped-flow vs continuous-flow

	Stopped-flow	**Continuous-flow**
Cryogenic separation of Xe	No	Yes
Xe in gas mix	>10%	1–3%
Laser powers	50–170	50–170
Typical ^{129}Xe polarisation	30%–95%	10%–50%
Xe production rates	100s ml/h	1000s ml/h
Low cell temp	[Rb] $\leq 10^{12}$ cm^{-3}	[Rb] 10^{12}–10^{14} cm^{-3}

Stopped-flow: Higher ^{129}Xe polarisations, lower Xe production rates

Continuous-flow: Lower ^{129}Xe polarisations, higher Xe production rates

Continuous-flow ^{129}Xe-Rb SEOP



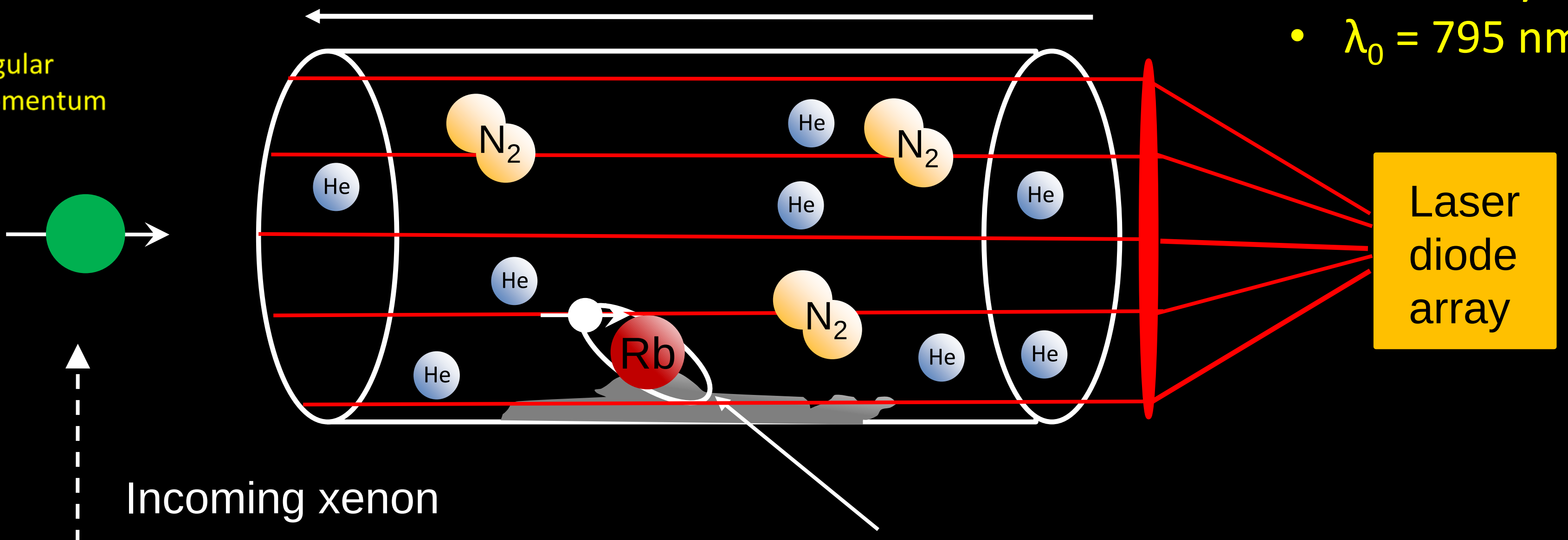
Rb optical pumping

1–3% Xe

Need to purify – cryogenic separation…

- Circularly polarised light
- σ^+ helicity
- λ_0 = 795 nm

B_0 = 10–50 G

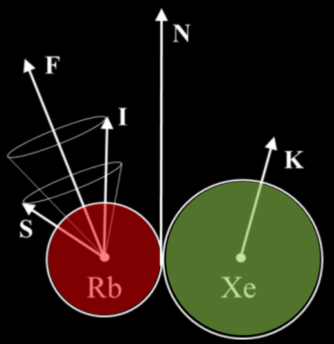


Incoming xenon

Bed of rubidium

T > 40°C
(313.15 K)

Rb vapour density curve



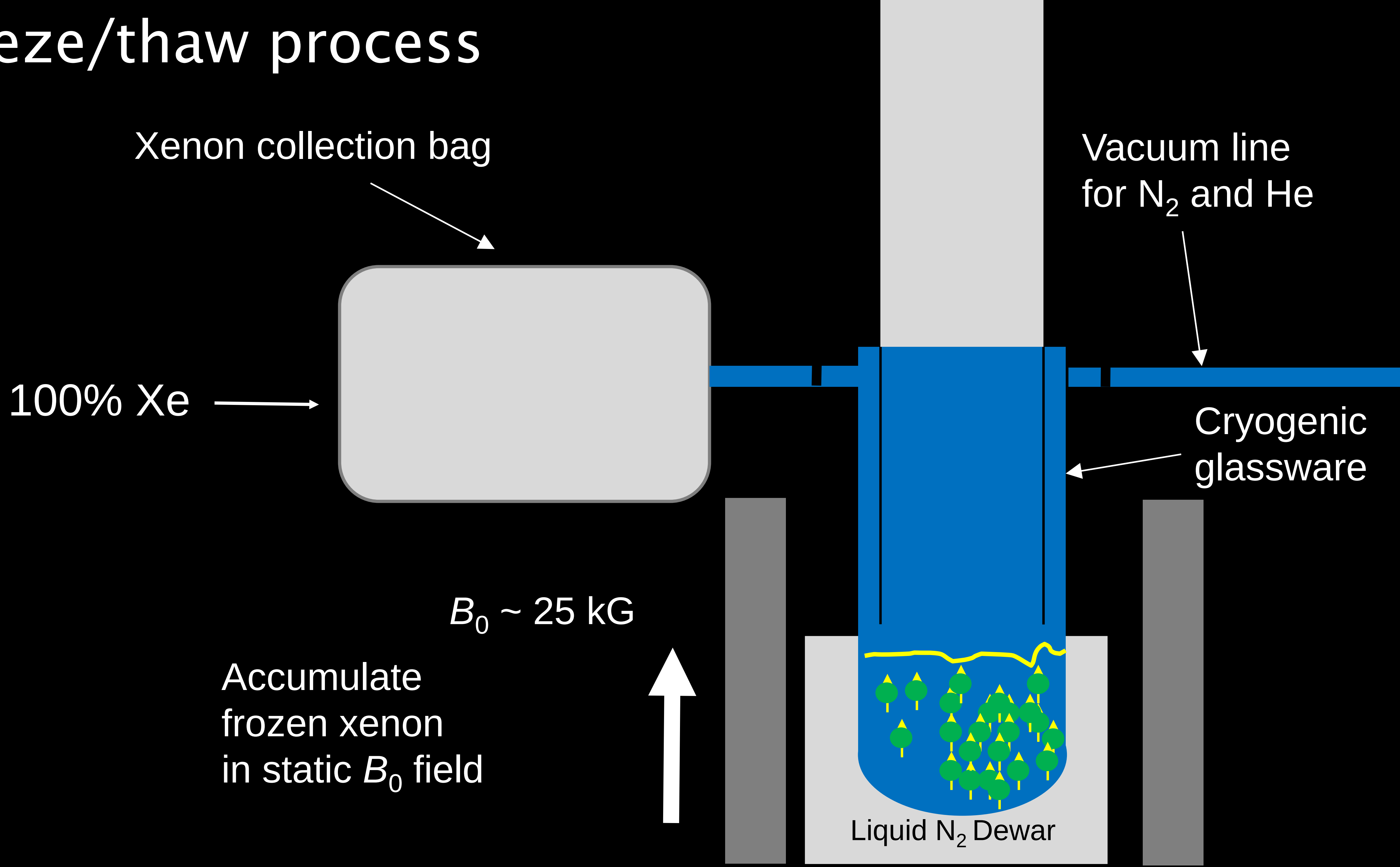
$$H = A\mathbf{I}\cdot\mathbf{S} + \gamma\mathbf{N}\cdot\mathbf{S} + \alpha\mathbf{K}\cdot\mathbf{S} + \cdots$$

High Rb–Xe spin-destruction cross section

^{129}Xe-Rb spin exchange

Path to cryogen

Freeze/thaw process

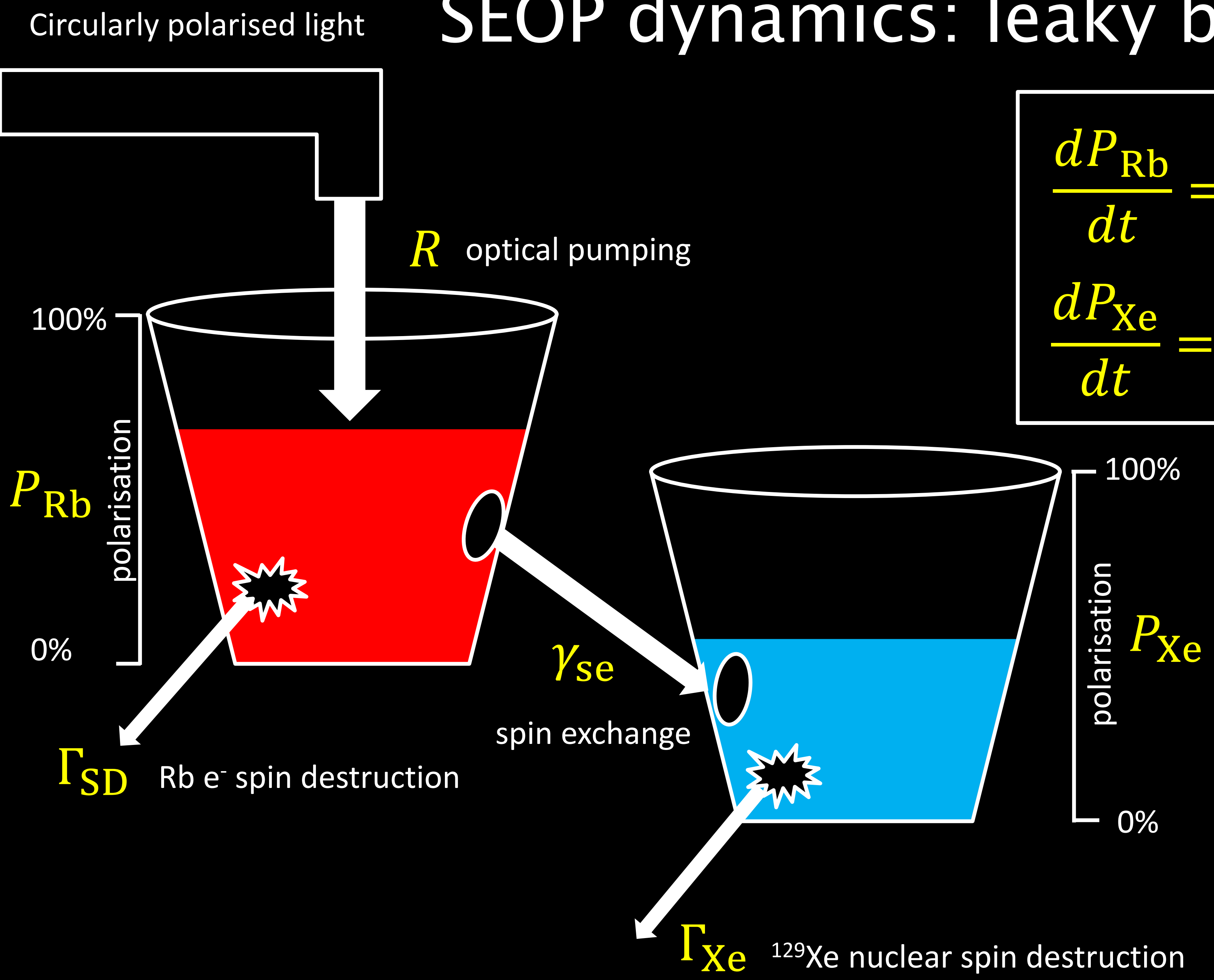


SEOP dynamics: leaky bucket model

$$\frac{dP_{\mathrm{Rb}}}{dt} = (1 - P_{\mathrm{Rb}})R - \Gamma_{\mathrm{SD}} P_{\mathrm{Rb}}$$

fill rate — leak rate

$$\frac{dP_{\mathrm{Xe}}}{dt} = \gamma_{\mathrm{se}}(P_{\mathrm{Rb}} - P_{\mathrm{Xe}}) - \Gamma_{\mathrm{Xe}} P_{\mathrm{Xe}}$$



P_{Rb} and P_{Xe} time dependence

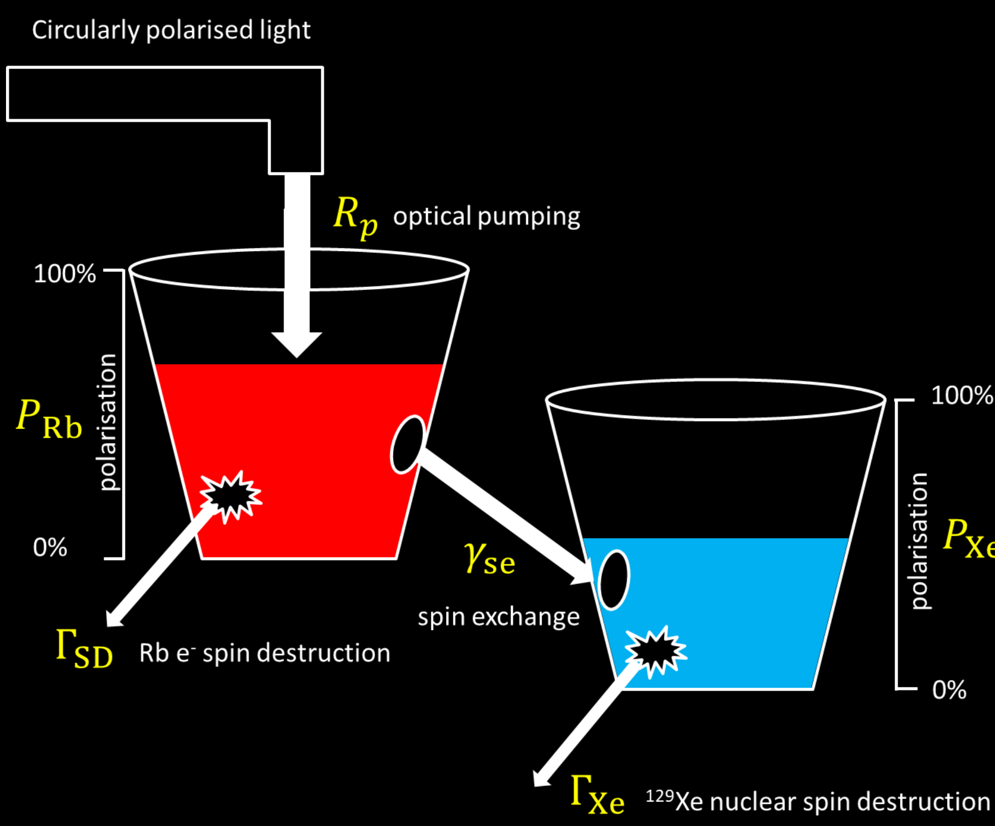
$$\frac{dP_{\mathrm{Rb}}}{dt} = (1 - P_{\mathrm{Rb}})R - \Gamma_{\mathrm{SD}} P_{\mathrm{Rb}}$$

$$\frac{dP_{\mathrm{Xe}}}{dt} = \gamma_{\mathrm{se}}(P_{\mathrm{Rb}} - P_{\mathrm{Xe}}) - \Gamma_{\mathrm{Xe}} P_{\mathrm{Xe}}$$

$$P_{\mathrm{Rb}}(t) = \frac{R}{R + \Gamma_{\mathrm{SD}}}\left(1 - e^{-(R+\Gamma_{\mathrm{SD}})t}\right)$$

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

Leaky bucket model



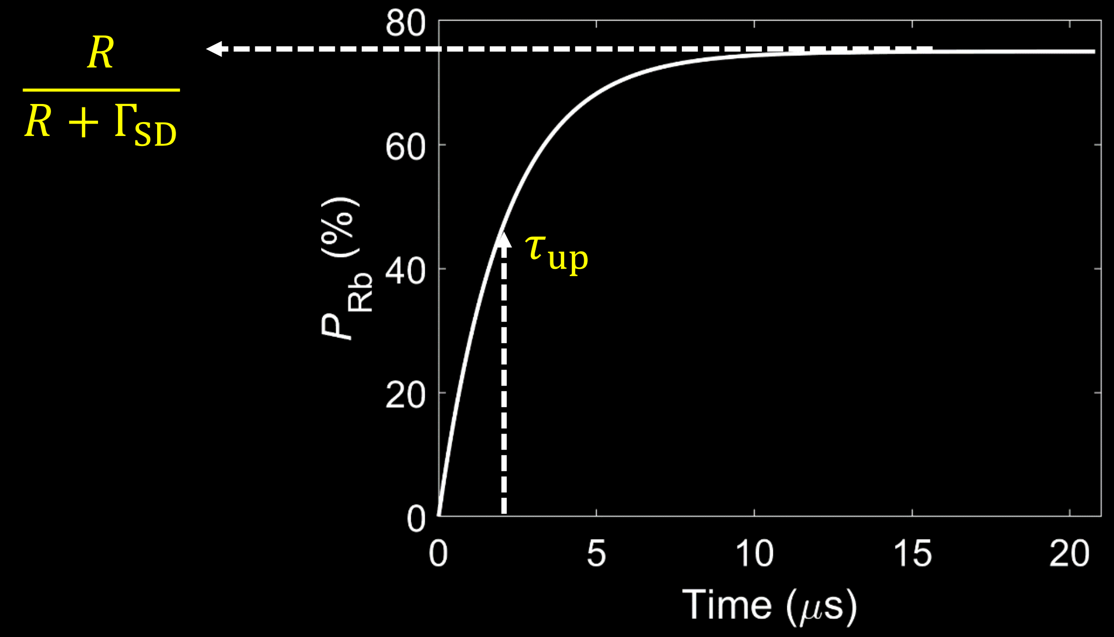
P_{Rb} and P_{Xe} steady-state and build up

Rb polarisation

$$P_{\text{Rb}}(t) = \frac{R}{R + \Gamma_{\text{SD}}}\left(1 - e^{-(R_p + \Gamma_{\text{SD}})t}\right)$$

Steady-state: $P_{\text{Rb}} = \dfrac{R}{R + \Gamma_{\text{SD}}}$

Spin-up time: $\tau_{\text{up}} = 1/(R_p + \Gamma_{\text{SD}}) \approx$ micro seconds

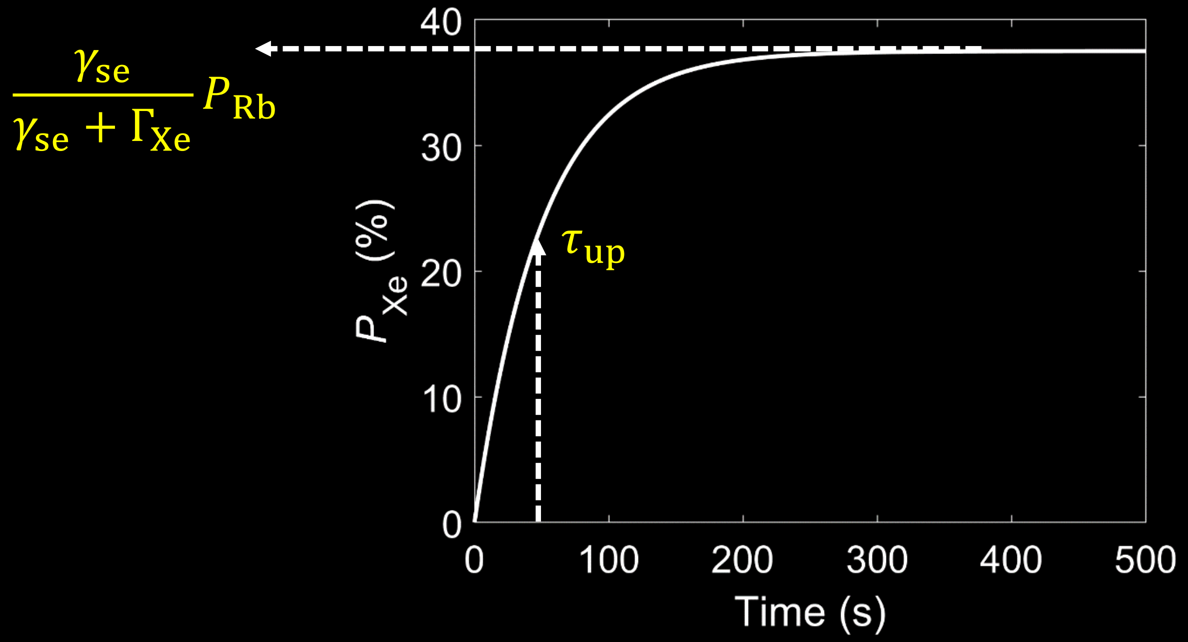


^{129}Xe polarisation

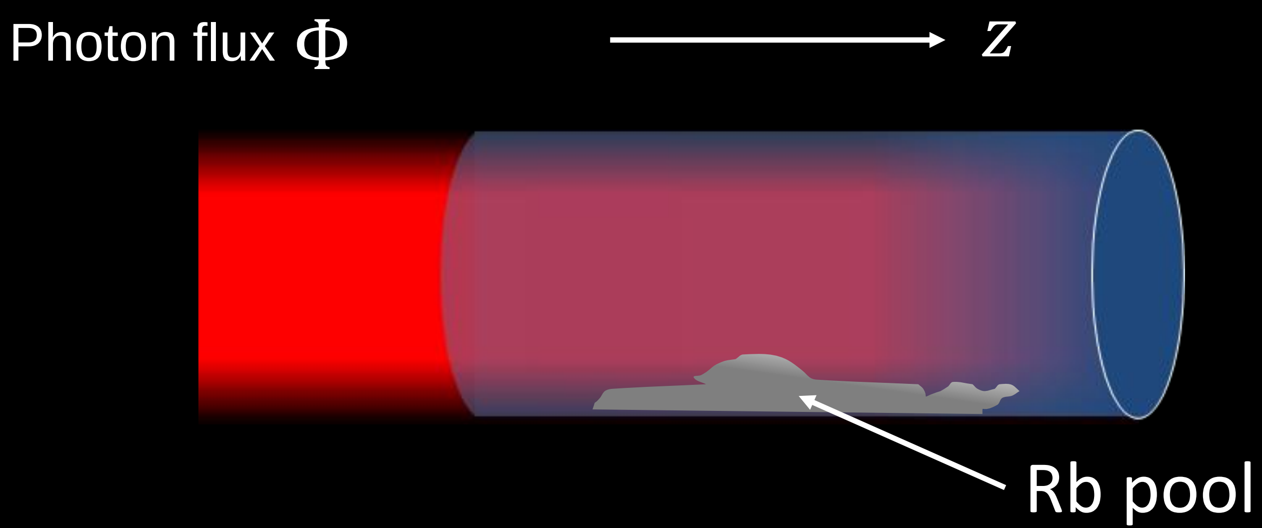
$$P_{\text{Xe}}(t) = \frac{\gamma_{\text{se}}}{\gamma_{\text{se}} + \Gamma_{\text{Xe}}} P_{\text{Rb}}(t)\left(1 - e^{-(\gamma_{\text{se}} + \Gamma_{\text{Xe}})t}\right)$$

Steady-state: $P_{\text{Xe}} = \dfrac{\gamma_{\text{se}}}{\gamma_{\text{se}} + \Gamma_{\text{Xe}}} P_{\text{Rb}}$

Spin-up time: $\tau_{\text{up}} = 1/(\gamma_{\text{se}} + \Gamma_{\text{Xe}}) \approx$ 10s of seconds



Light propagation: spatial dependence



$$\frac{\partial \Phi(\nu,z)}{\partial z} = -\underbrace{\lambda^{-1}(\nu,z)}\Phi(\nu,z)$$

λ = absorption length per photon

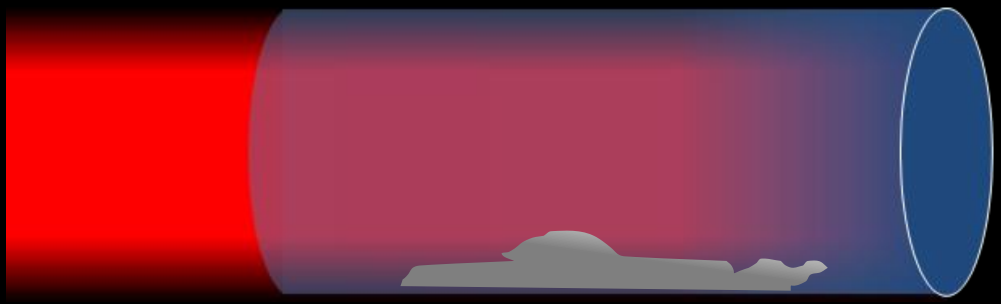
$$\lambda^{-1}(\nu,z) = [\mathrm{Rb}]\sigma(\nu)[1 - P_{\mathrm{Rb}}(z)]$$

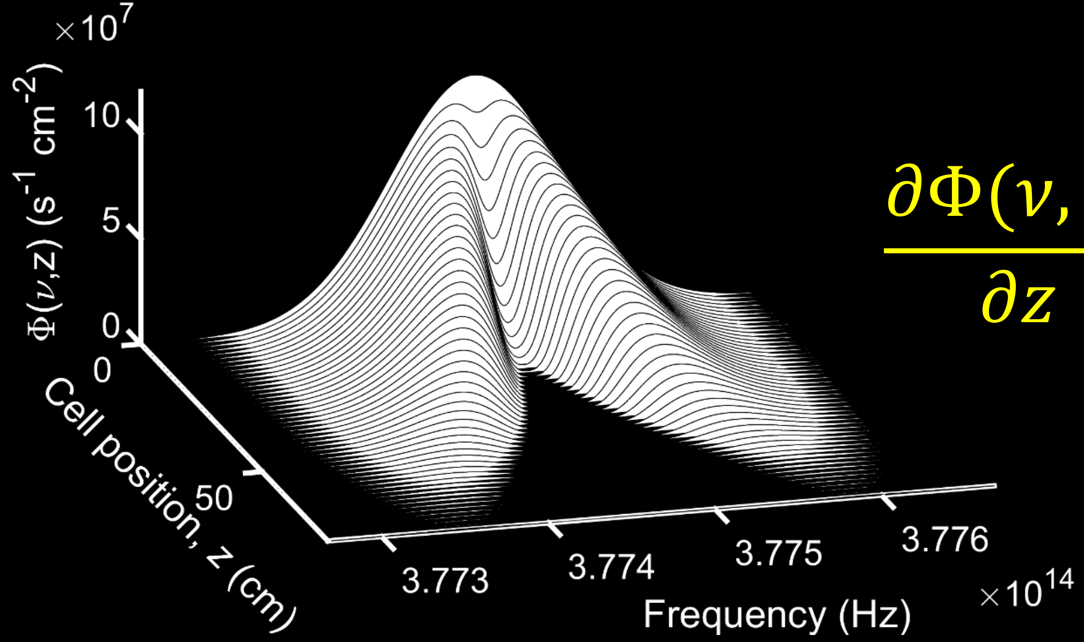
- $[\mathrm{Rb}]$: Rb vapour density
- $\sigma(\nu)$: Rb D1 line absorption profile
- $P_{\mathrm{Rb}}(z)$: Rb polarisation

$$\frac{\partial \Phi(\nu,z)}{\partial z} = -[\mathrm{Rb}]\sigma(\nu)[1 - P_{\mathrm{Rb}}(z)]\Phi(\nu,z)$$

Absorption profile over cell length

Photon flux Φ $\longrightarrow z$





Incident flux

$$\Phi(\nu,0) = \Phi_0 \exp\left[-4\ln 2\,\frac{(\nu-\nu_l)^2}{\Delta\nu_l^2}\right]$$ Gaussian

Absorption

$$\sigma(\nu) = \sigma_0 \frac{\Delta\nu_a/2\pi}{(\nu-\nu_a)^2 - \left(\frac{\Delta\nu_a}{2}\right)^2}$$ Lorentzian

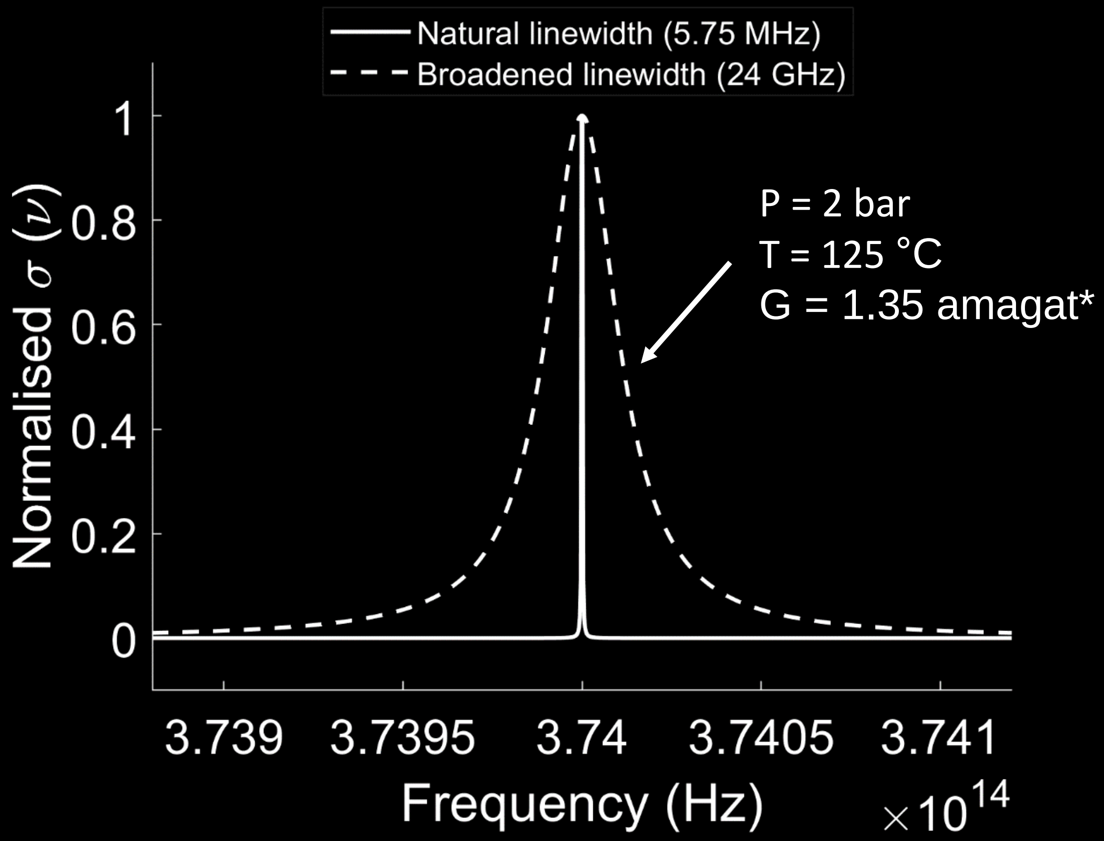
$$\frac{\partial\Phi(\nu,z)}{\partial z} = -[\mathrm{Rb}]\sigma(\nu)[1-P_{\mathrm{Rb}}(z)]\Phi(\nu,z)$$

$\Delta\nu_l = 142$ GHz (0.30 nm)

$\Delta\nu_a = 24$ GHz (0.05 nm)

Role of buffer gases N_2 and He

Pressure broadening of D_1 line

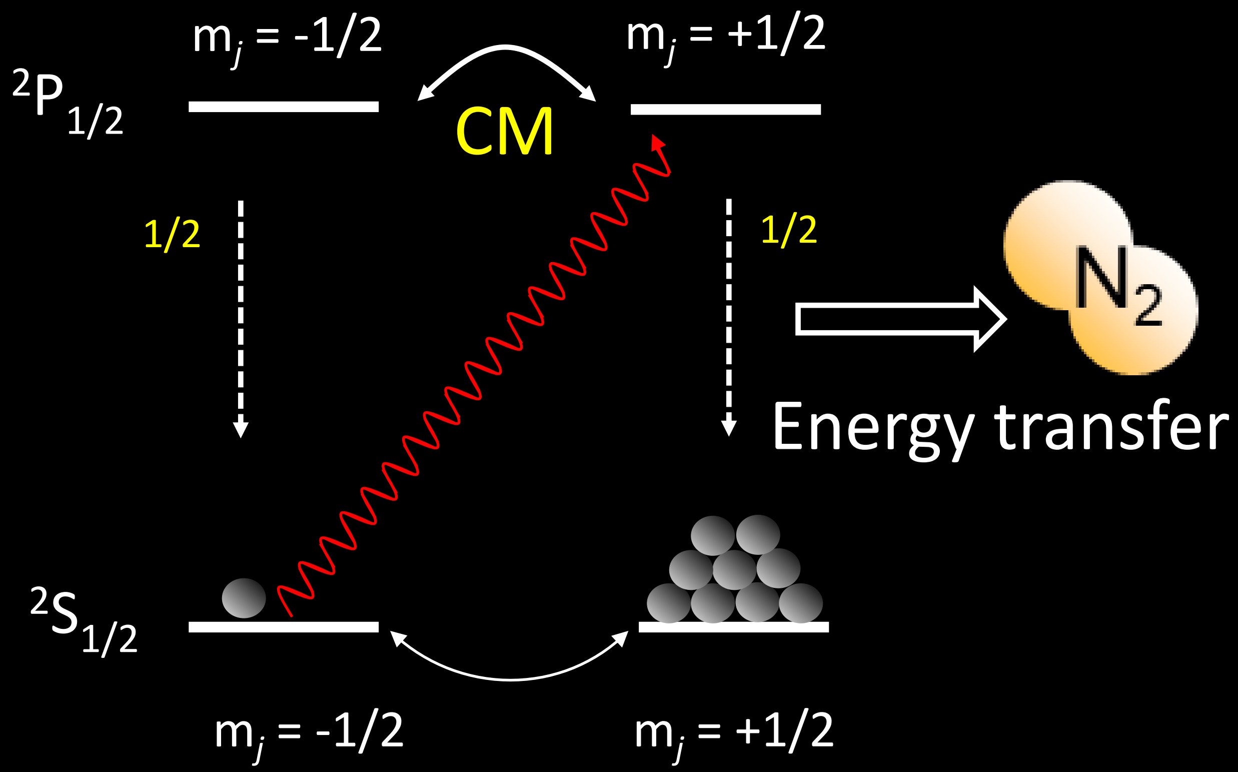


$He/N_2 : \Delta\nu_a \sim 18$ GHz/amagat

Romalis, PRA(56), 1997

*1 amagat = gas number density at STP (T = 0°C and P = 1 atm)

Non-radiative decay (quenching)

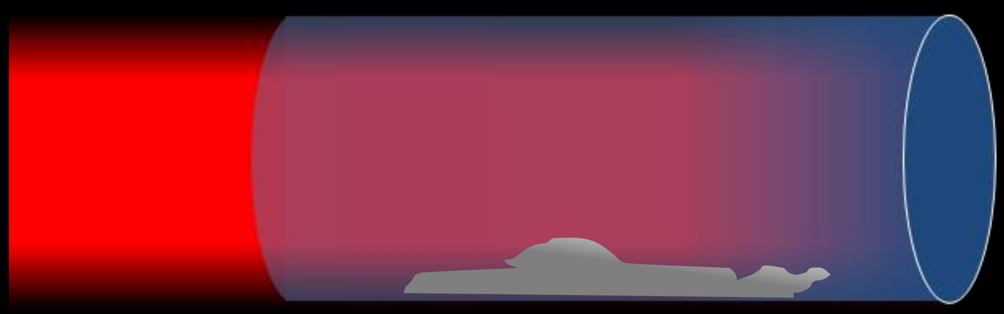


CM = collisional mixing

Wagshul and Chupp, PRA(40), 1989

Light propagation and optical pumping

Photon flux Φ $\longrightarrow z$



$$R(z) = \int \Phi(\nu, z)\sigma(\nu)d\nu$$

Optical pumping rate

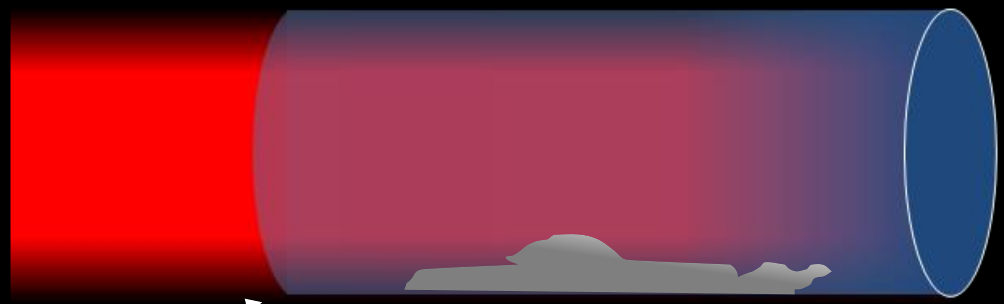
$$\frac{\partial\Phi(\nu, z)}{\partial z} = -[\mathrm{Rb}]\sigma(\nu)[1 - P_{\mathrm{Rb}}(z)]\Phi(\nu, z)$$

$$\frac{d\Phi(z)}{dz} = \int \frac{\partial\Phi(z, \nu)}{\partial z} d\nu = -[\mathrm{Rb}][1 - P_{\mathrm{Rb}}(z)]\boxed{\int \Phi(\nu, z)\sigma(\nu)d\nu}$$

$$\boxed{\frac{d\Phi(z)}{dz} = -[\mathrm{Rb}][1 - P_{\mathrm{Rb}}(z)]R(z)}$$

Relates photon attenuation with pumping rate

Optical pumping rate vs cell length



$$\frac{d\Phi(z)}{dz} = -[\mathrm{Rb}][1 - P_{\mathrm{Rb}}(z)]R(z)$$

$$R_0(\nu, 0) = \int_{-\infty}^{\infty} \sigma(\nu)\Phi(\nu) = \alpha\Phi$$

$$\alpha = \frac{2\sqrt{\pi \ln 2}\, r_e f \lambda_l^3 w'(r,s)}{hc\Delta\lambda n_p}$$

Appelt, PRA(52), 1998

$$\frac{dR(z)}{dz} = -[\mathrm{Rb}]\alpha[1 - P_{\mathrm{Rb}}(z)]R(z)$$

$$W\big(f(x)\big) = x, \text{for } f(x) = xe^x$$

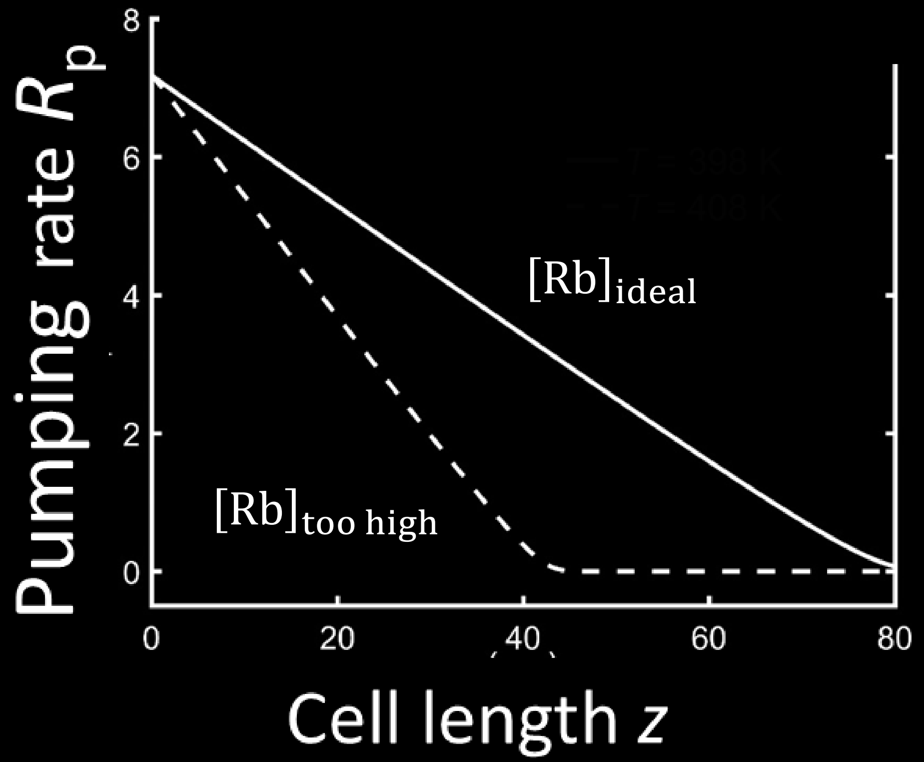
Product log function

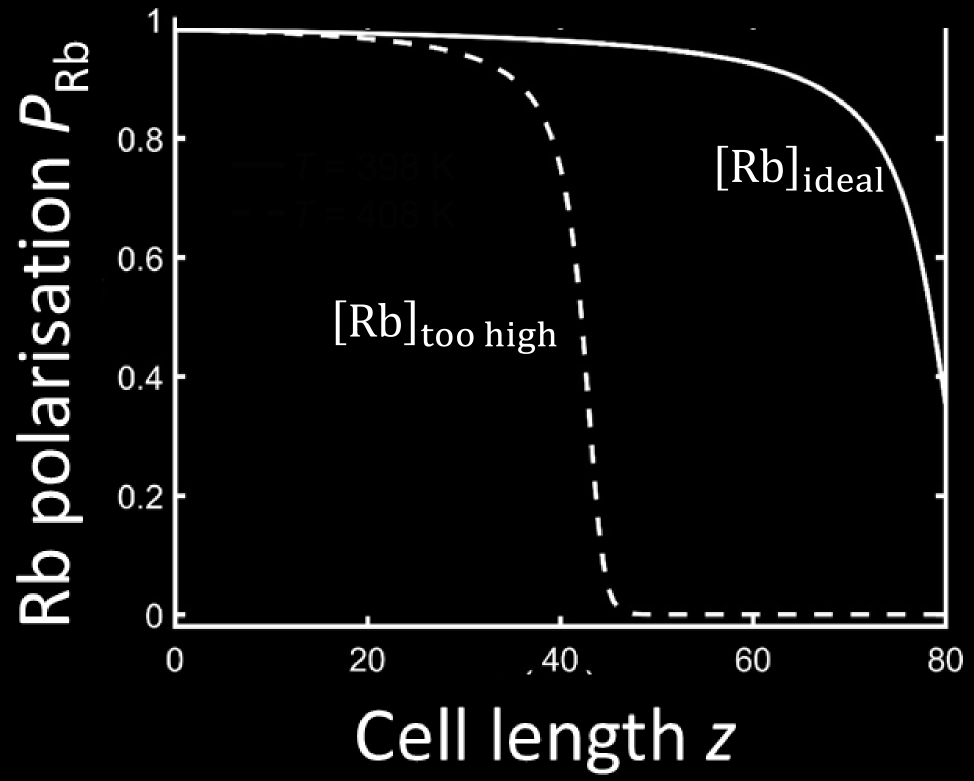
$$R(z) = \Gamma_{SD} W\left(\frac{R_0}{\Gamma_{SD}} \exp\left(\frac{R_0}{\Gamma_{SD}} - \alpha[\mathrm{Rb}]z\right)\right)$$

Optimisation tools: Light propagation, R_p and P_Rb

$$R(z) = \Gamma_{SD} W\left(\frac{R_0}{\Gamma_{SD}} \exp\left(\frac{R_0}{\Gamma_{SD}} - \alpha[\mathrm{Rb}]z\right)\right)$$

$$P_{\mathrm{Rb}}(z) = \frac{R(z)}{R(z) + \Gamma_{SD}}$$

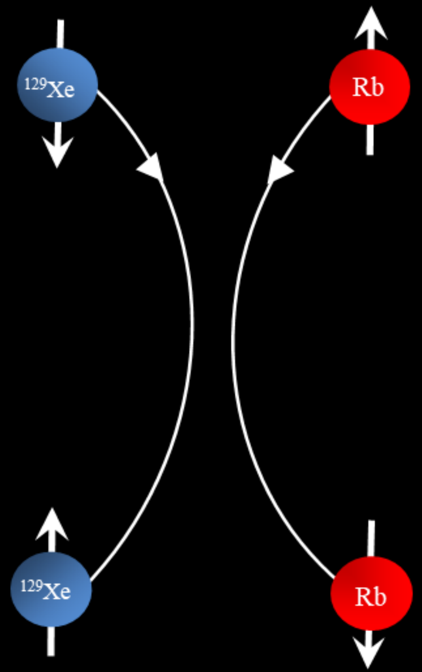




Solid white line necessary for high average P_Rb over the cell volume

^{129}Xe-Rb spin exchange

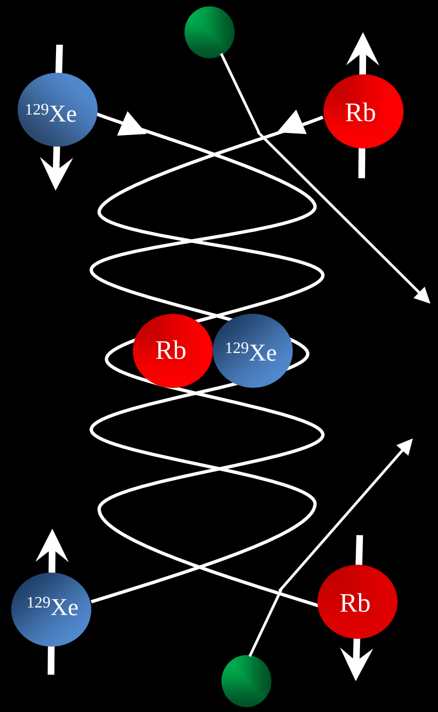
Two-body collisional exchange



- Simple two-body cross section
- Independent of P_{Rb} and gas composition

Three-body vdW molecular exchange

3rd body {N_2, He, Xe}



- Complicated cross section
- Depends on P_{Rb} and gas composition

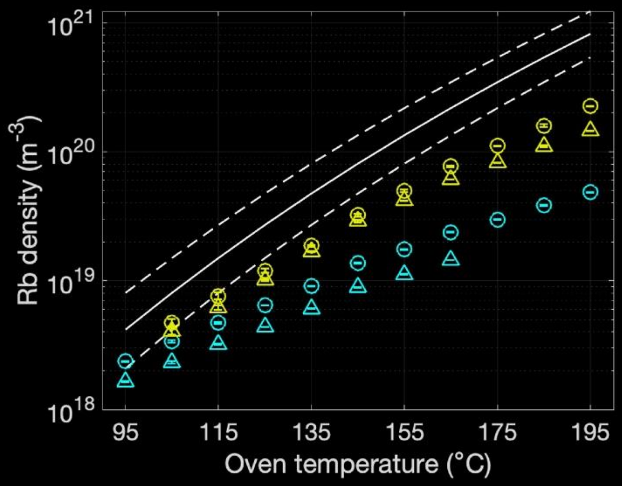
^{129}Xe-Rb spin exchange

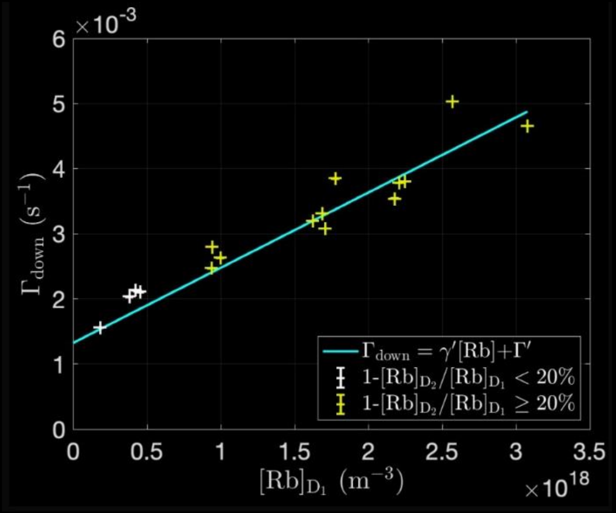
Two-body collisional exchange

$$\gamma_{se}^{bc} = \langle \sigma v \rangle \, [\text{Rb}]$$

Binary spin-exchange cross section

Ball et al, Molecules(8), 2023





Factor-2 lower [Rb] than standard saturation curves

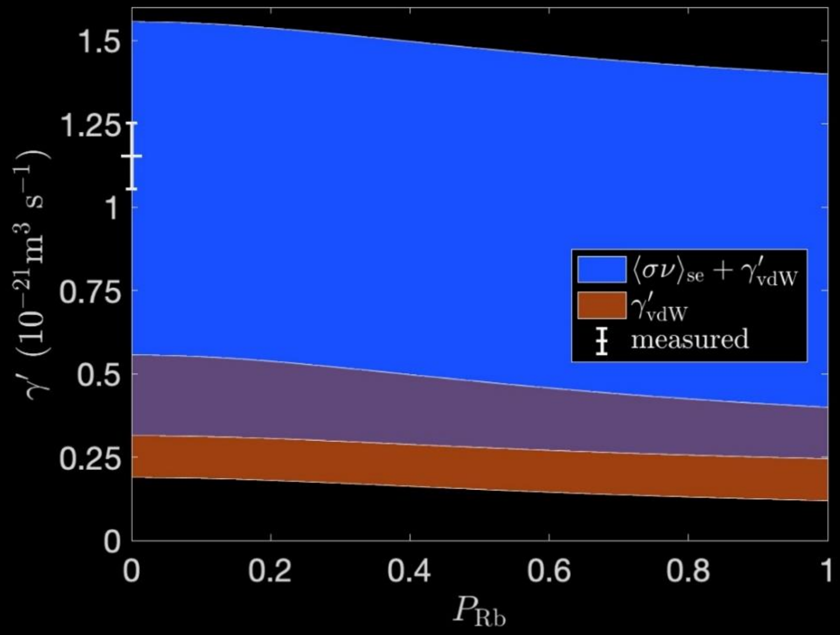
$$\gamma' = \langle \sigma v \rangle + \gamma'_{vdW} = 1.2 \times 10^{-21} \ \text{cm}^3\text{s}^{-1}$$

Three-body vdW molecular exchange

$$\gamma_{se}^{vdw} = \frac{1}{2T_K}\left(\frac{\omega_{sr}\tau}{x}\right)^2 \sum_i \eta_i \left(\frac{1 + q_i(\omega_{h,i}\tau)^2/[I_i]^2}{1 + (\omega_{h,i}\tau)^2}\right) = \gamma'_{vdW}[\text{Rb}]$$

$\tau =$ molecular lifetime ⟵ Gas composition dependent

$q_i = 1 + \epsilon(I, P_{\text{Rb}})$ ⟵ Nuclear slowing down factor



Ball et al, Molecules(8), 2023

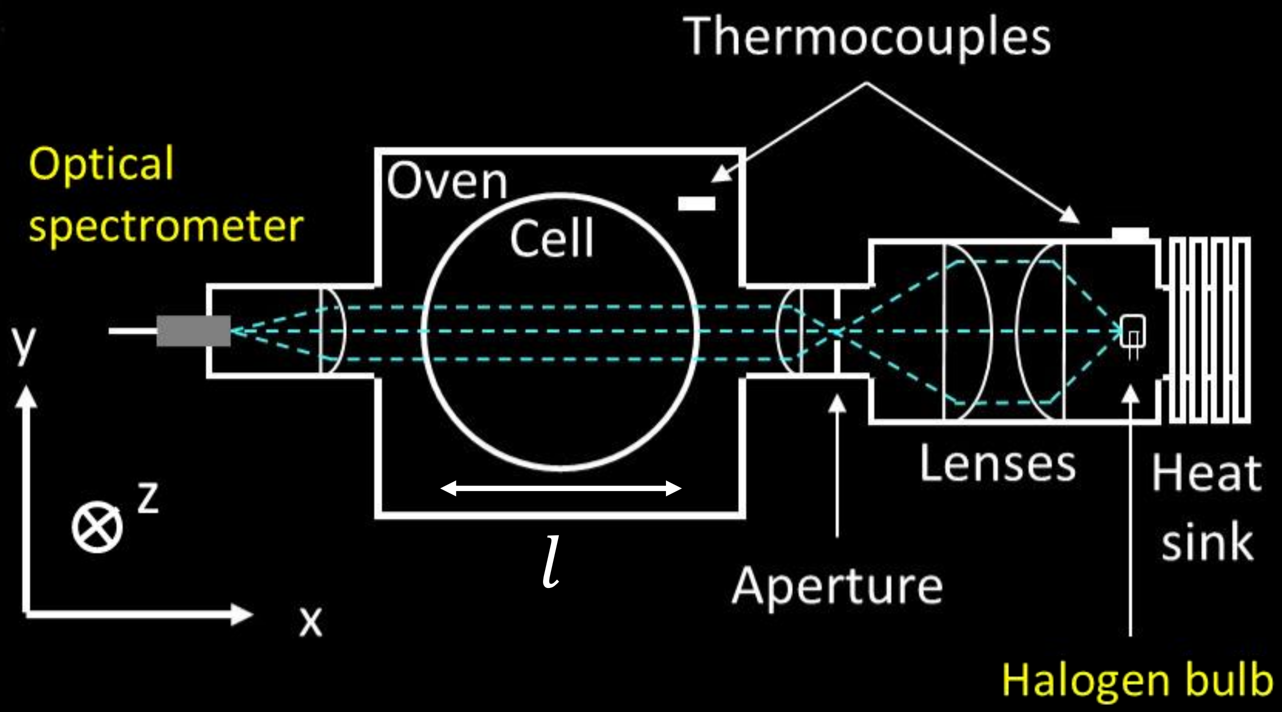
Literature: $\langle \sigma v \rangle = 0.1 \times 10^{-21}$ to 1×10^{-21} cm^{3}s^{-1}

Factor-10 range in values...

Measuring Rb vapour density

Atomic Absorption spectroscopy

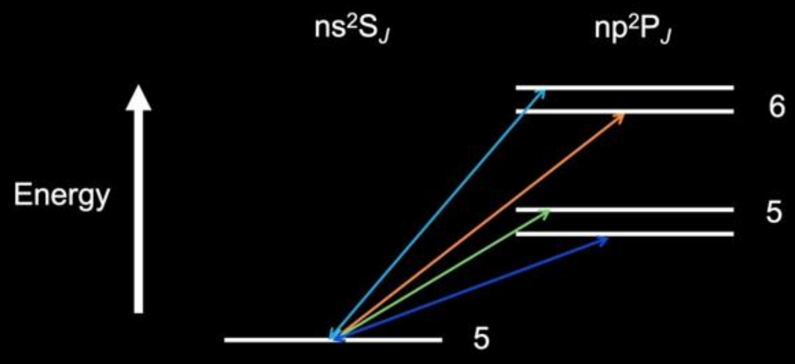
Experimental Set up



Beer-Lambert law

$$I(\nu) = I_0 e^{-[\mathrm{Rb}]\sigma(\nu) l}$$

Absorption cross section

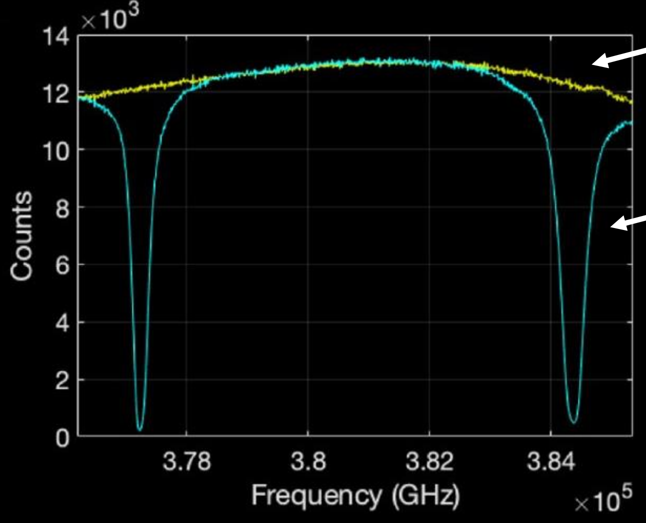


Transition	Wavelength (nm)	Oscillator strength, *f*
$5^2S_{1/2} \rightarrow 6^2P_{1/2}$	422	3.87×10^{-3}
$5^2S_{1/2} \rightarrow 6^2P_{3/2}$	420	9.46×10^{-3}
$5^2S_{1/2} \rightarrow 5^2P_{1/2}$ (D_1)	795	3.422×10^{-1}
$5^2S_{1/2} \rightarrow 5^2P_{3/2}$ (D_2)	780	6.957×10^{-1}

Measuring Rb vapour density

Atomic Absorption spectroscopy

$$I = I_0 e^{-[\mathrm{Rb}]\sigma(\nu) l}$$



Absorbance ↓

Amplitude ↙

$$S(\nu) = \ln\left(\frac{I_0}{I}\right) = A L(\nu)$$

Normalised Lorentzian

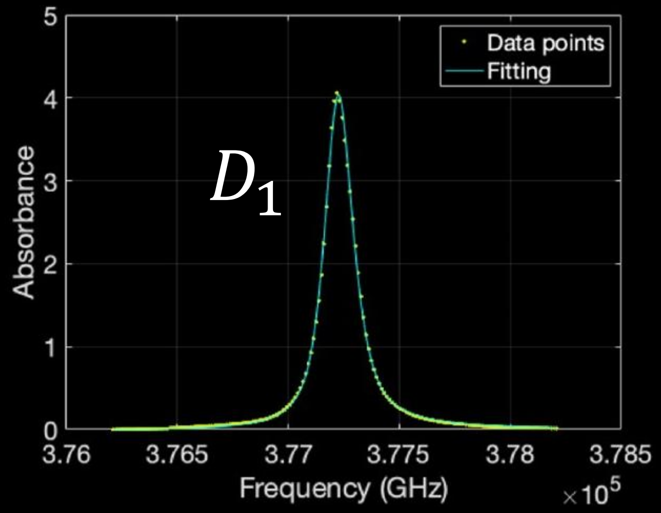
Transition oscillator strength

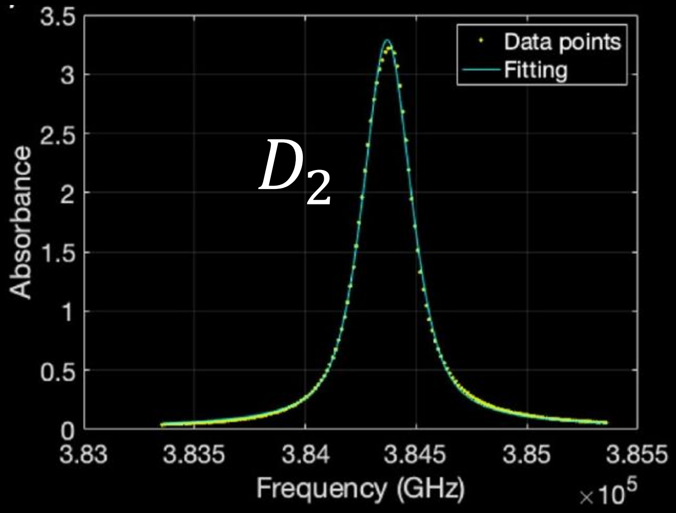
r_0: electron radius

c: light speed

$$[\mathrm{Rb}] = \frac{1}{\sigma_0 l}\int S(\nu) d\nu$$

$$\sigma_0 = \int \sigma(\nu) d\nu = \pi r_0 c f$$





$$[\mathrm{Rb}] = \frac{A}{\pi r_0 c f l}$$

Optimisation tools: ^{129}Xe polarisation

mean Rb polarisation over cell volume

Xe cell residency time

$$P_{\mathrm{Xe}}(t_{\mathrm{res}}) = \frac{\gamma_{\mathrm{se}}}{\gamma_{\mathrm{se}} + \Gamma_{\mathrm{Xe}}} \langle P_{\mathrm{Rb}} \rangle \left[1 - e^{-t_{\mathrm{res}}/\tau_{\mathrm{up}}}\right]$$

$\gamma_{se} = \gamma_{se}^{bc} + \gamma_{se}^{vdW}$

^{129}Xe spin-exchange rate

^{129}Xe spin-destruction rate

^{129}Xe spin-up time

$$\gamma_{\mathrm{se}} \propto [\mathrm{Rb}]$$

$$\Gamma_{\mathrm{Xe}} = 1/T_1$$

$$\tau_{\mathrm{up}} = 1/(\gamma_{\mathrm{se}} + \Gamma_{\mathrm{Xe}})$$

For high P_{Xe} :

$\gamma_{\mathrm{se}} > 1/T_1$ | High $\langle P_{\mathrm{Rb}} \rangle$ | $t_{\mathrm{res}} > \tau_{\mathrm{up}}$

Continuous-flow polariser optimisation

Rationale for a large cell

- Long cell maximises absorption of incident photons at lower [Rb]

- Laser heating/Rb runaway less likely

- At high gas flow rate Q and lower [Rb] $t_{\mathrm{res}} > \tau_{\mathrm{up}}$ for large volume cell

$$t_{\mathrm{res}} = \frac{[\mathrm{G}]V_{\mathrm{cell}}}{Q} \qquad \tau_{\mathrm{up}} \propto 1/[\mathrm{Rb}]$$

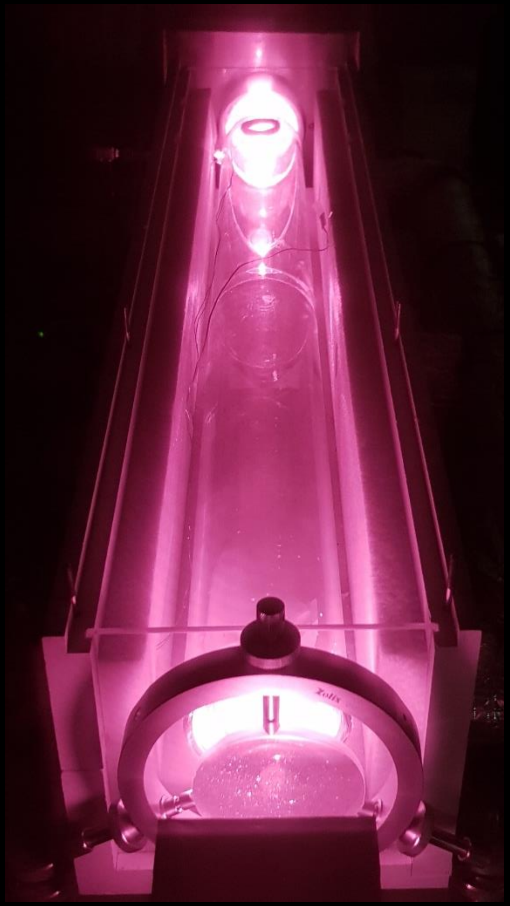
High P_{Xe} at rapid Xe production rates

Polariser design: large-volume optical cell

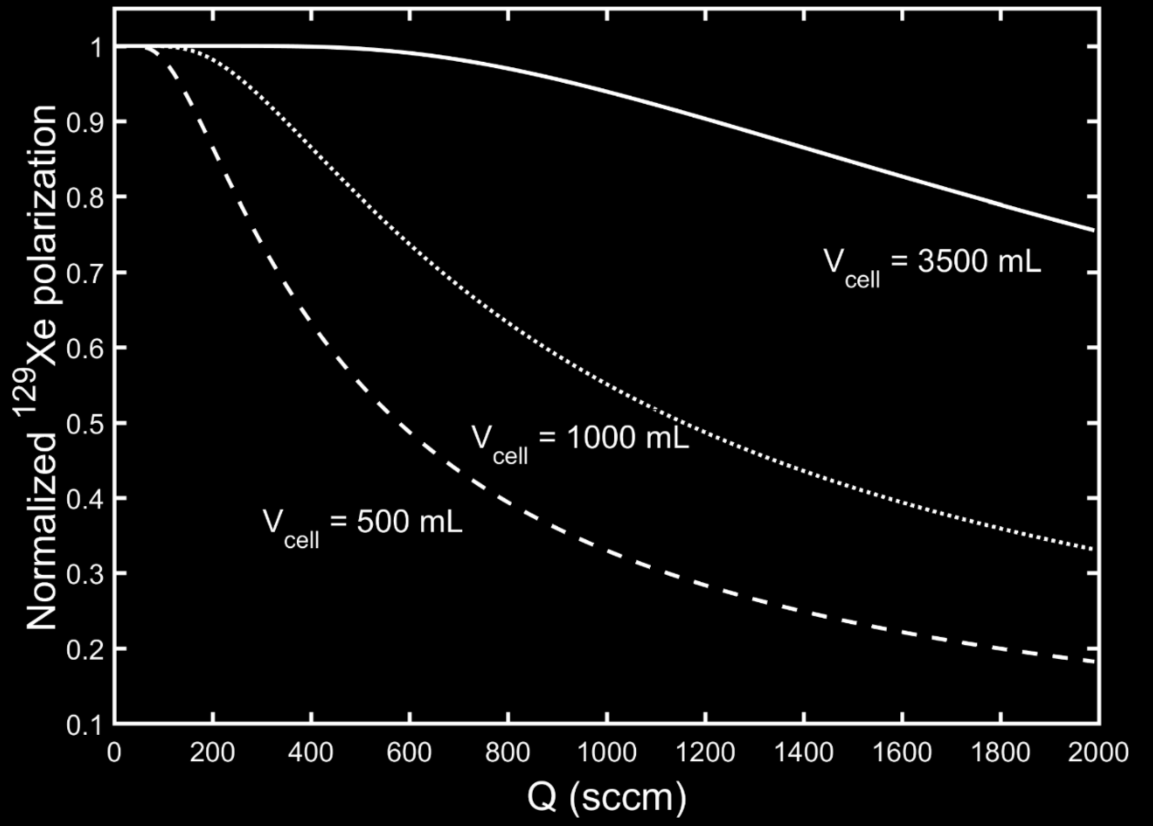
$$P_{Xe}(t_{res}) = P_0(1 - e^{-t_{res}/\tau_{up}})$$

$$P_{Xe}(Q) = P_0(1 - e^{-[G]V_{cell}/Q\tau_{up}})$$

$$t_{res} = \frac{[G]V_{cell}}{Q}$$



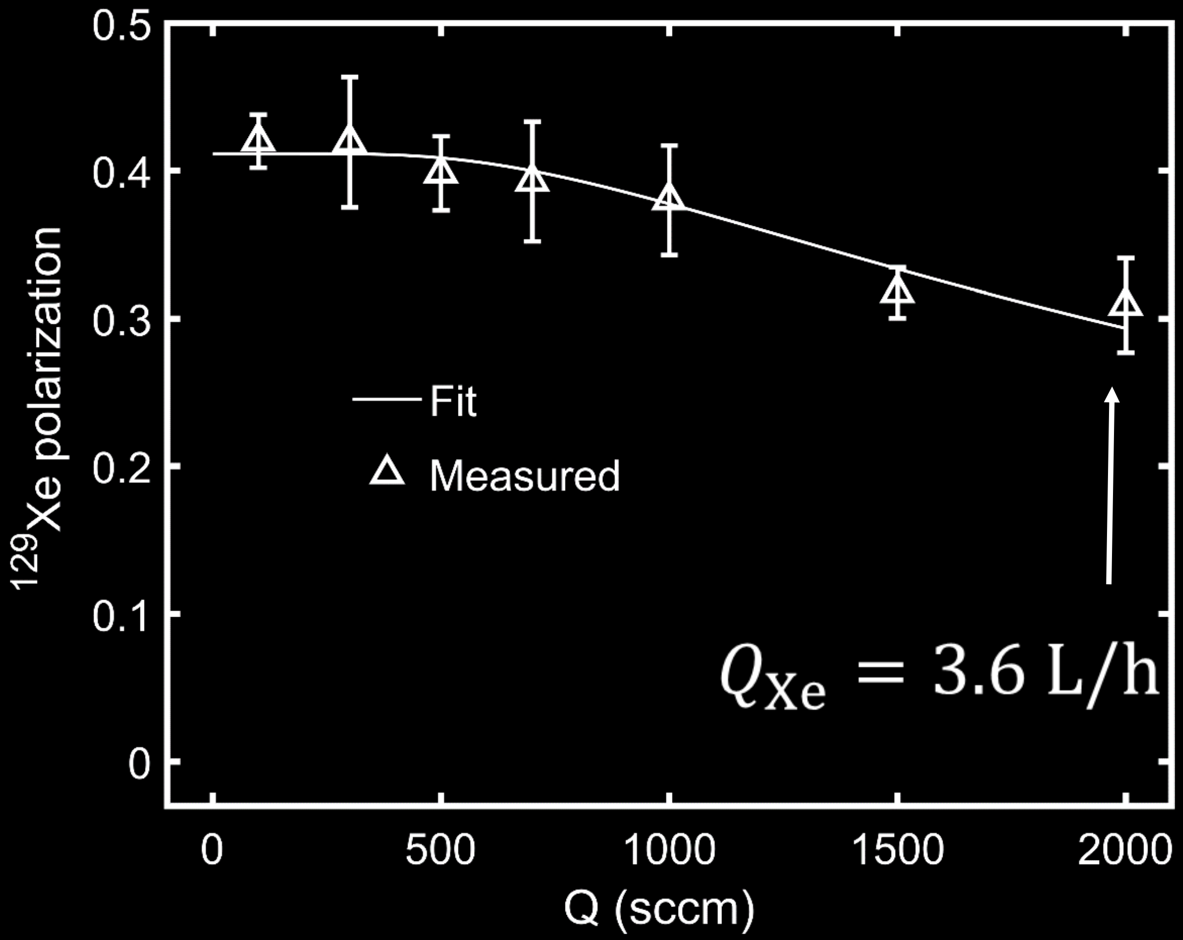
Cell dimensions:
- Length = 80cm
- Diameter = 7.5 cm
- Volume = 3530 mL



Simulated:

- T_{cell} = 398 K
- P_{cell} = 1.25 bar
- [G] = 0.85 amg

^{129}Xe polariser optimisation



Optimised parameters:

- T_{cell} = 398 K
- P_{cell} = 1.25 bar
- 3% Xe gas mix
- [G] = 0.85 amg
- V_{cell} = 3530 mL

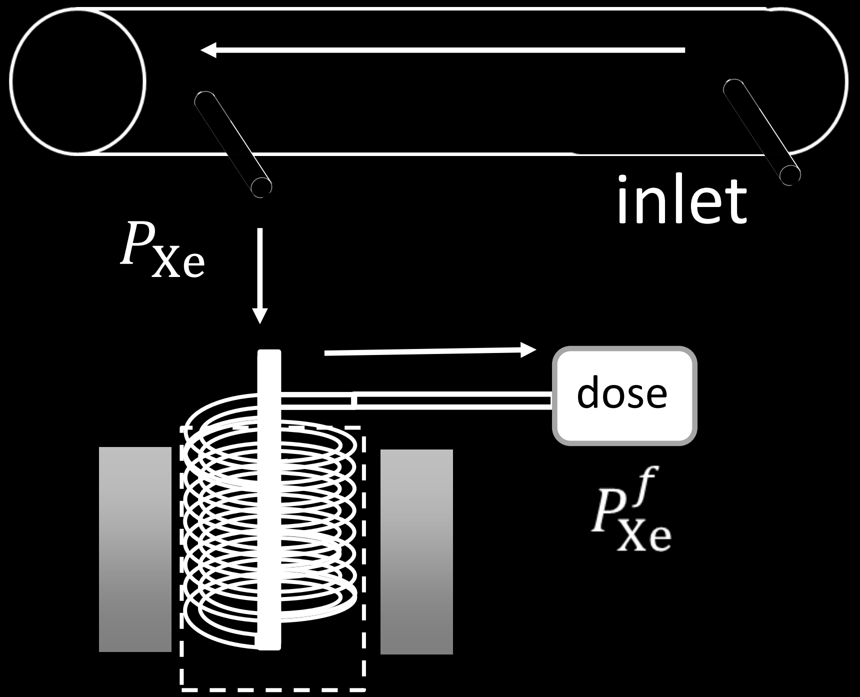
$Q = 2000$ sccm ⇨ $t_{res} = 90$ s

$P_{Xe}(Q) = P_0(1 - e^{[G]V_{cell}/Q\tau_{up}})$ ⇨ $\tau_{up} = 71$ s ⇨ $P(Q = 2000) \approx 0.75 P_0$

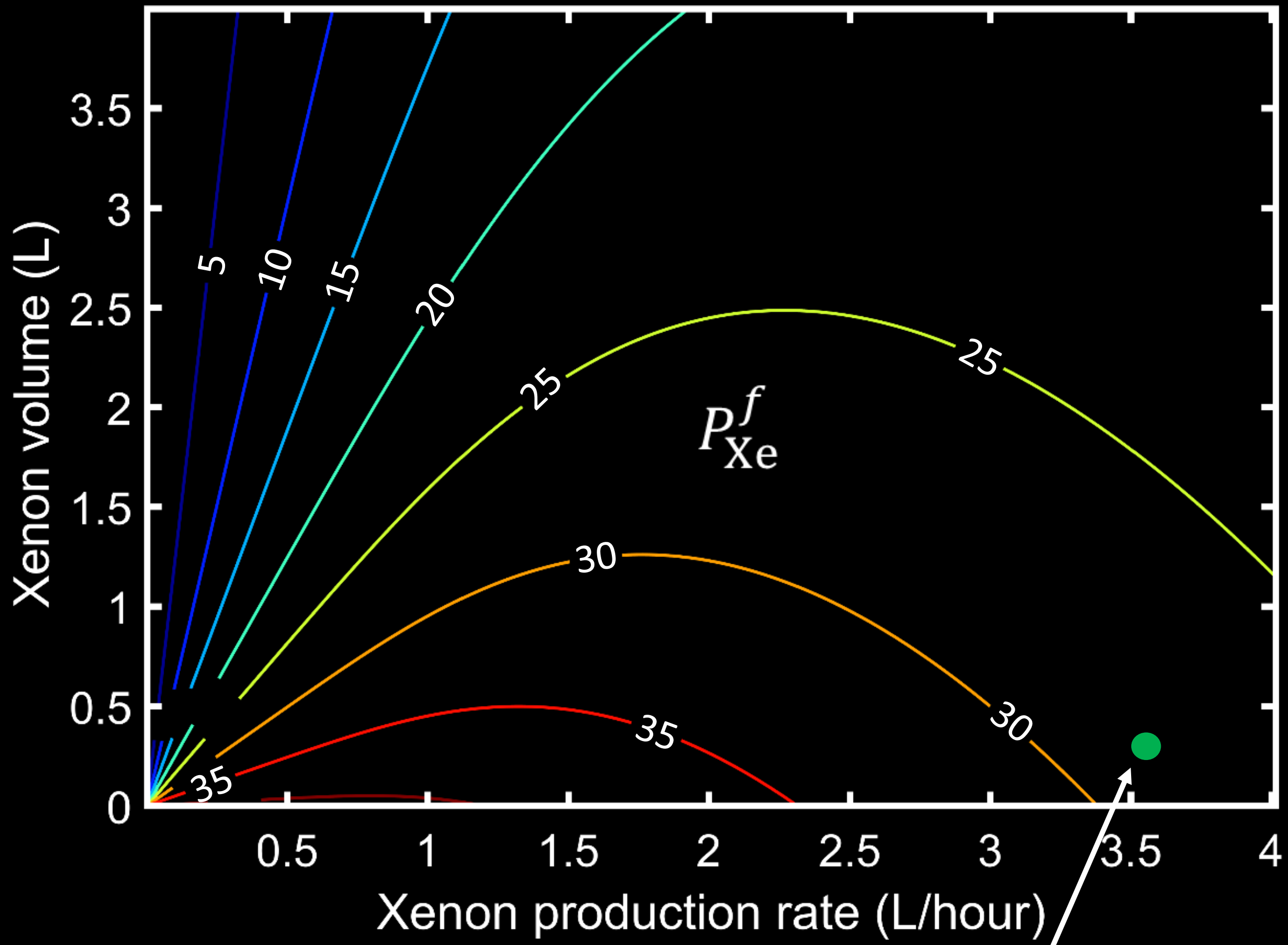
Norquay et al., PRL(121), 2018

^{129}Xe production map

$\tau_{\mathrm{up}} = 71\ \mathrm{s}$



Frozen Xe, $T_1^S = 90\ \mathrm{min}$*



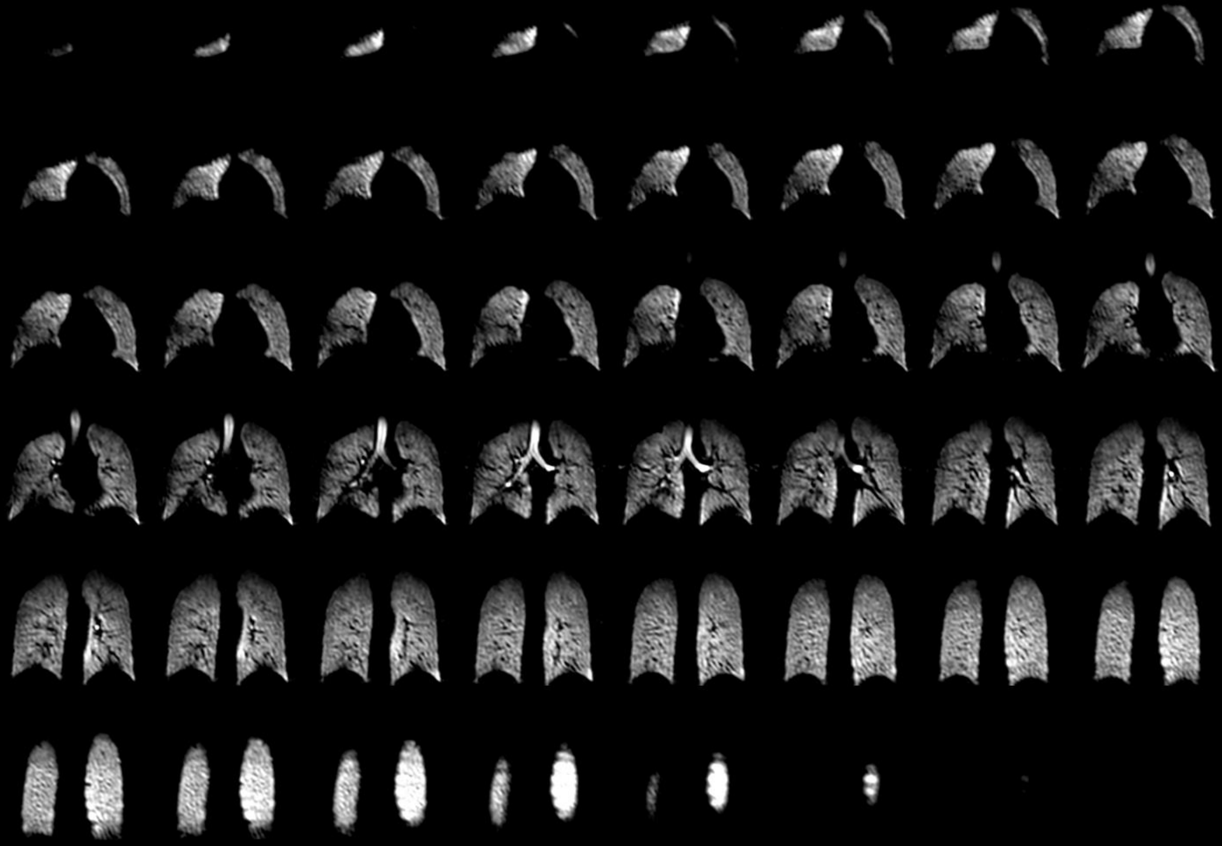
$$P_{\mathrm{Xe}}^{f}(Q, t_a) = P_{\mathrm{Xe}}(Q)\left[1 - e^{-t_a/T_1^S}\right]T_1^S/t_a$$

300 mL, P_{Xe} ~30% in 5 min

*Norquay et al., J Appl Phys(13), 2013

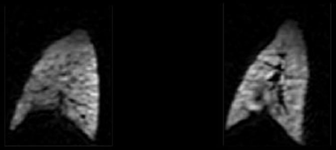
Norquay et al., Phys Rev Lett(121), 2018

3D ^{129}Xe human lung imaging with enriched Xe



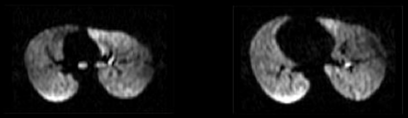
coronal

Enriched Xe:
1L dose generated
~20 min



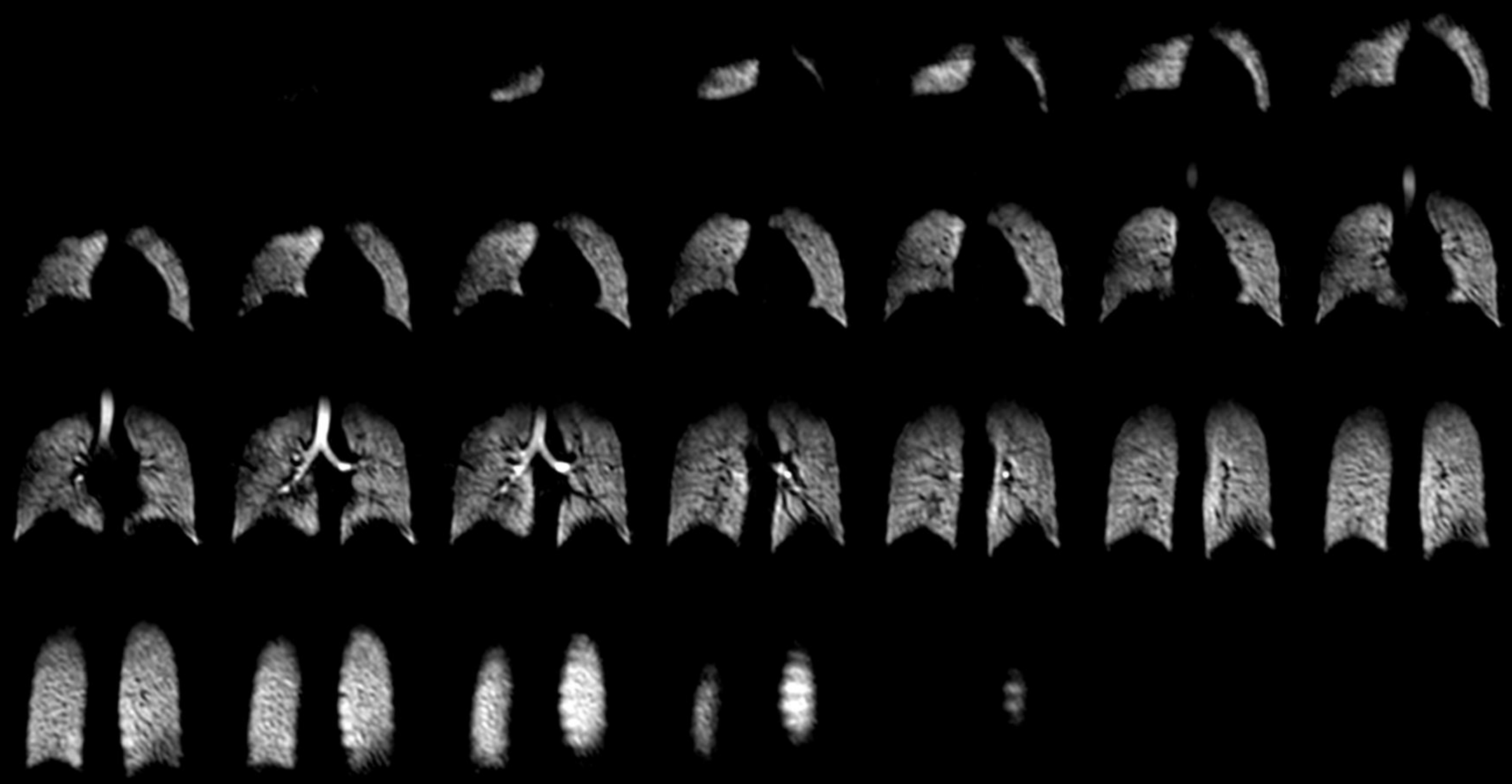
sagittal

Isotropic resolution: **4.2 mm^3**



axial

3D ^{129}Xe human lung imaging with NA Xe



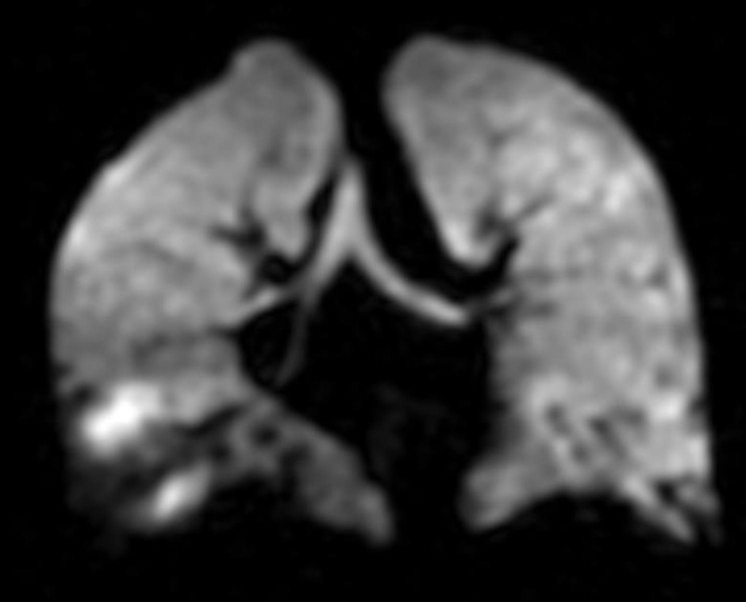
$25/scan
1/8 cost of enriched Xe

1L natural abundance Xe in 20 min

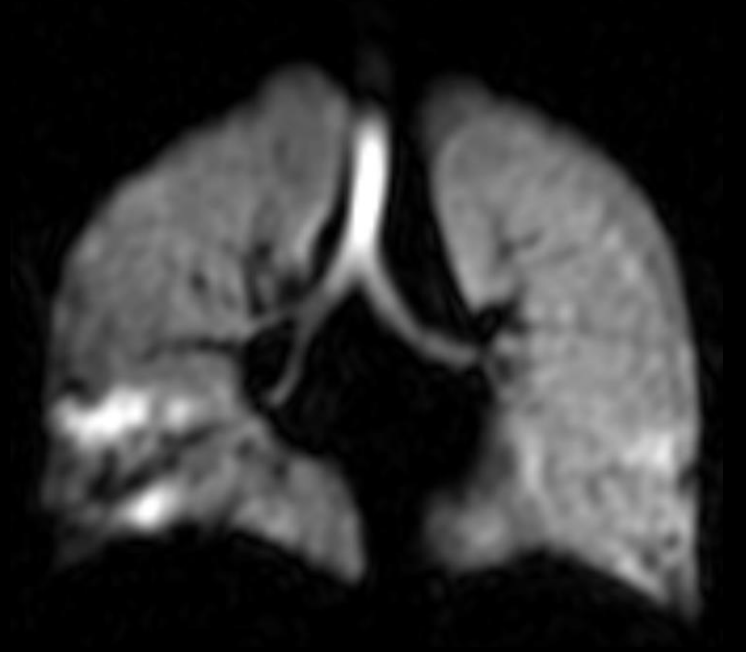
Stewart., et al. MRM (74), 2015

Low-dose paediatric clinical ^{129}Xe lung imaging

^{3}He (120 mL)



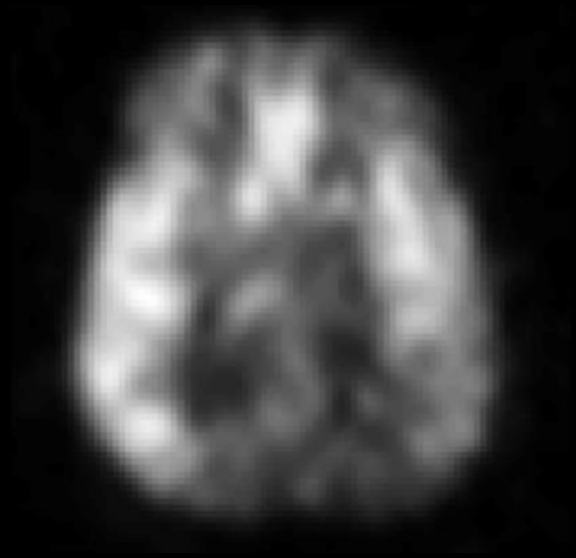
^{129}Xe (250 ml)



250 mL ^{129}Xe dose generated < 5 min

Dissolved ^{129}Xe applications beyond the lungs

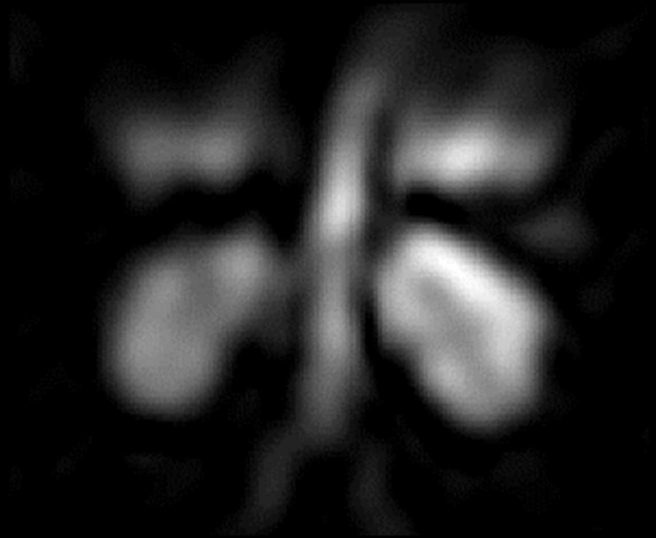
1.5 T



^{129}Xe in the brain

Rao et al., Radiology(286), 2018

3.0 T



Bottom of lungs

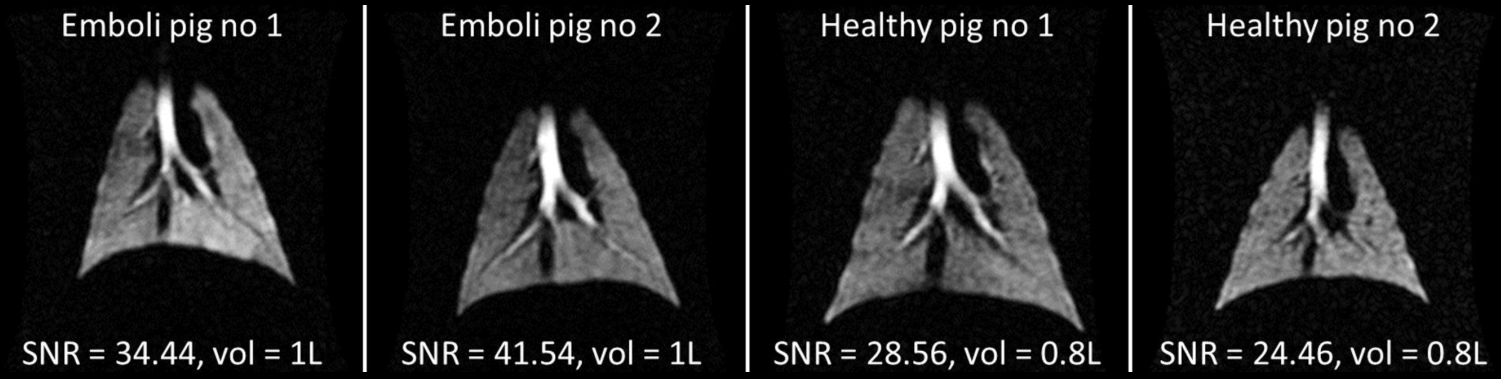
Kidneys

^{129}Xe in the kidneys

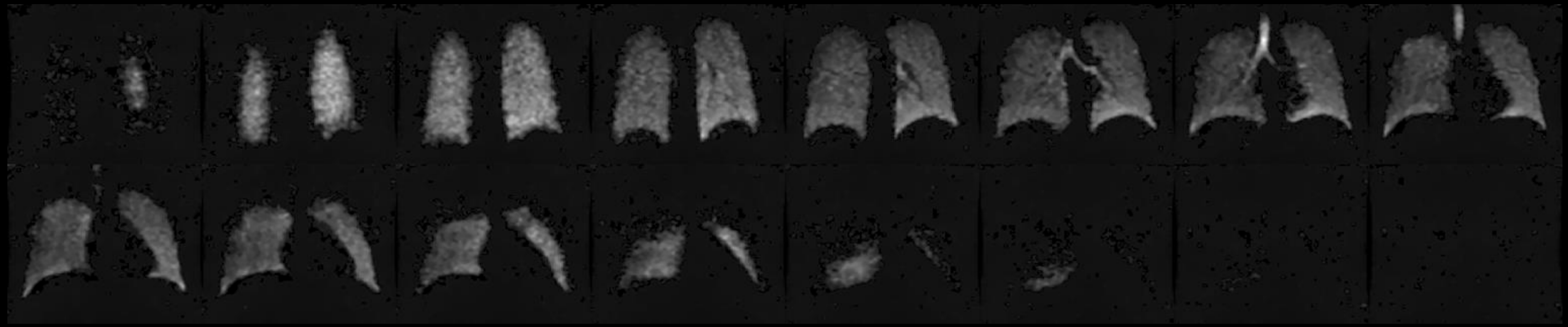
Chacon-Caldera et al., MRM(83) 2019

Imaging ^{129}Xe straight from the cell

3% xenon on tap, no cryogenic accumulation



PORCINE LUNGS



HUMAN LUNGS

Vaeggemose et al., Proc ISMRM, p4506, 2023

Summary

Optimising a polariser system is a complex, multi-parameter problem

[Rb] density critical parameter for process efficiency – generally lower in practice compared with theory

Currently wide range of published exchange rate constants – more work needed

Current unknowns

Cause of SEOP cell deterioration over time

Mechanism driving cell T_1 variations – orientation to B_0 field? Cell T_1 temperature dependence?

Thanks for your attention

Questions?