

# Enhancing the sensitivity of CASPEr using Dynamic Nuclear Polarization



Janos Adam

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Boston University

APS DPF Northeastern



Alfred P. Sloan  
FOUNDATION



HEISING - SIMONS  
FOUNDATION

**BOSTON  
UNIVERSITY**

SIMONS  
FOUNDATION

# Quick review of CASPEr

Three possible couplings:

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

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

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

$$H_e \propto a \vec{\sigma} \cdot \vec{E}^*$$

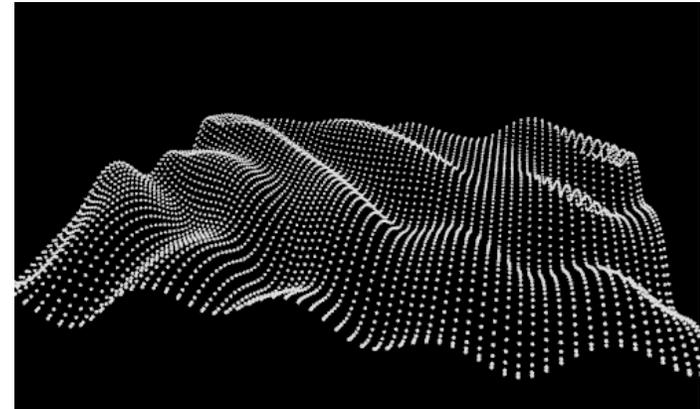
Ultralight axion field:

$$a(t) = a_0 \cos \omega_a t$$

$\omega_a = m_a c^2 / \hbar \rightarrow$  ALP Compton frequency

$\rho_{\text{DM}} \propto a_0^2 \rightarrow$  dark matter density

$$H_e \propto \vec{\sigma} \cdot (a_0 \vec{E}^*) \cos \omega_a t$$



**This interaction resembles a Nuclear Magnetic Resonance Hamiltonian!**

# NMR and how we use it

$$H_{\text{NMR}} = \vec{\sigma} \cdot \vec{B}$$

Static magnetic field:  $B_0$

$$H_{\text{NMR}} = \vec{\sigma} \cdot \vec{B}_0 + \vec{\sigma} \cdot \vec{B}_1 \cos \omega_0 t$$

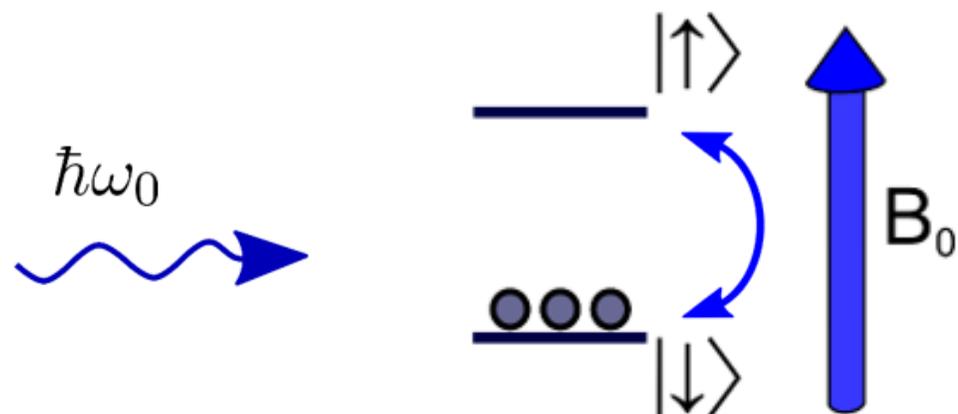
Oscillating perpendicular field:  $B_1$

Now, back to the axion Hamiltonian:

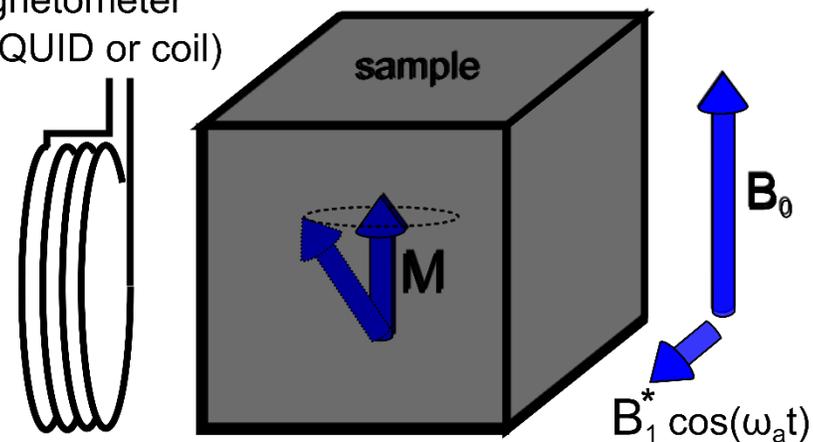
$$H_e \propto \vec{\sigma} \cdot (a_0 \vec{E}^*) \cos \omega_a t$$

And, the experimental realization is

$$H_{\text{CASPER}} = \vec{\sigma} \cdot \vec{B}_1^* \cos \omega_a t$$



magnetometer  
(eg, SQUID or coil)



**Nuclear spin interacts with the effective magnetic field – like in a NMR.**

# Extension of CASPEr – Phase 2

Expected sensitivity of our experiment in phase 1

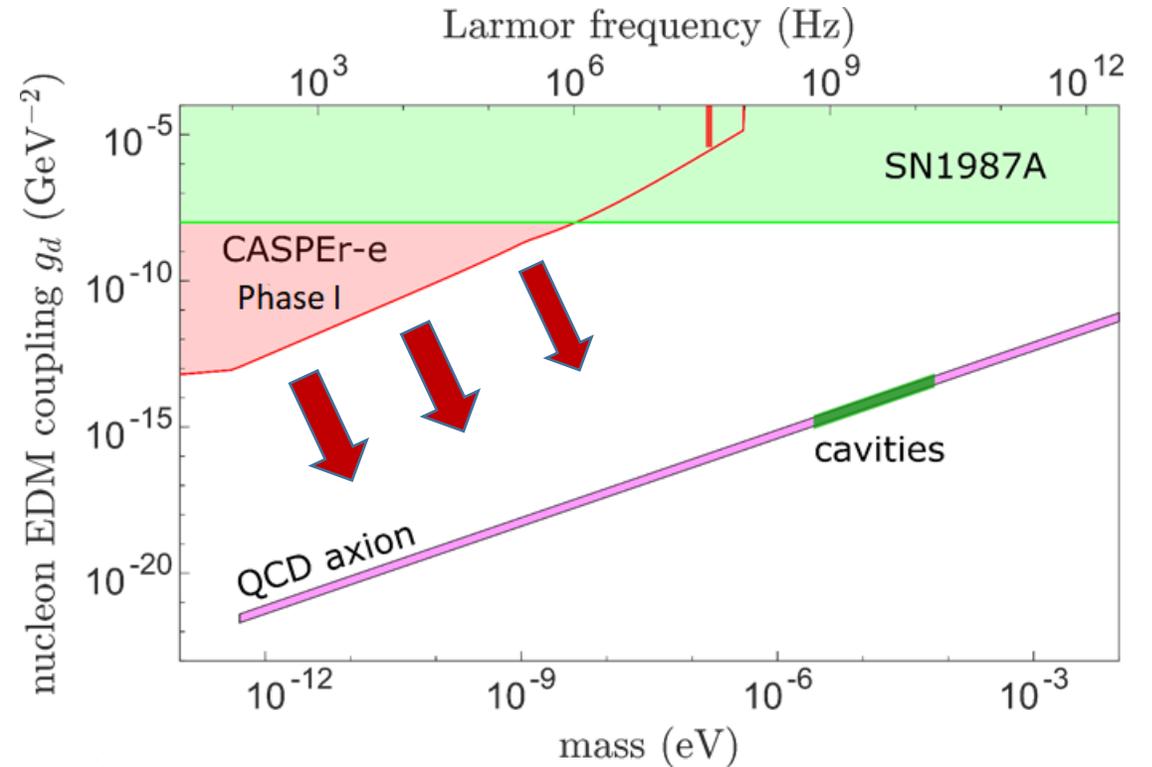
Effective interaction:

$$H_{\text{CASPEr}} = \vec{\sigma} \cdot \vec{B}_1^* \cos \omega_a t$$

1) Maximize  $\vec{B}_1^* = g_d a_0 \vec{E}^*$

2) Maximize spin polarization

Spin polarization  $p = \frac{n_\uparrow - n_\downarrow}{n_\uparrow + n_\downarrow} \sim \frac{\mu B}{k_B T} \sim 10^{-4}$



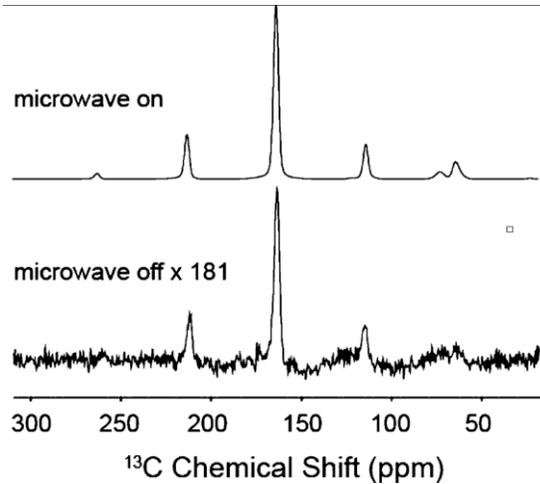
**Polarization is the key to increase sensitivity**

# How to increase polarization?

## Dynamic Nuclear Polarization (DNP)

DNP is transmitting polarization from electrons to nuclei

Common in chemistry/biology : eg  $^{13}\text{C}$



[Acc. Chem. Res. 2013, 46, 9, 1933-1941]

**BUT...**

Our sample doesn't have unpaired electron spins

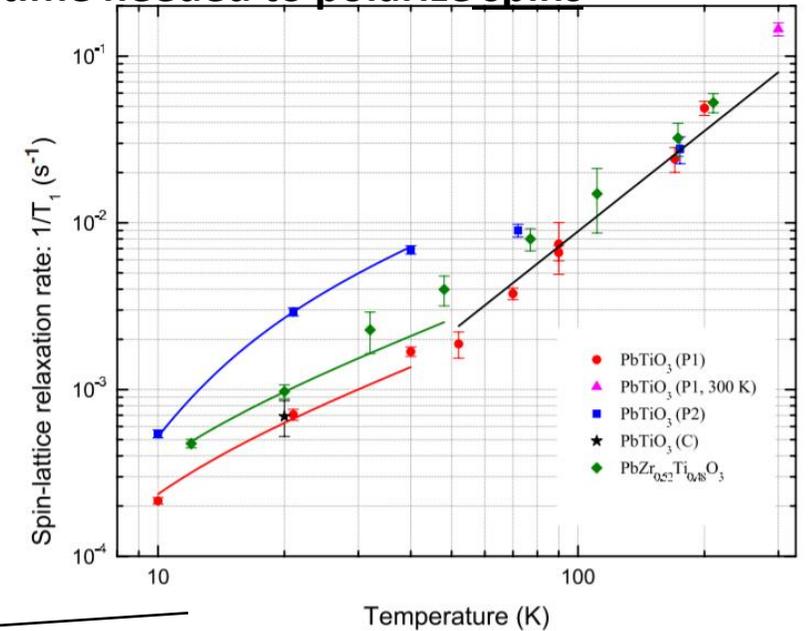
say, by a factor of 100

**Decrease temperature to 40 mK**

$$p \sim \mu_B / k_B T$$

**BUT...**

T1 (spin-lattice relaxation) becomes long (~ hours)  
Too much time needed to polarize spins



[Phys. Rev. A 77, 022102 (2008)]

**By shining laser we can**

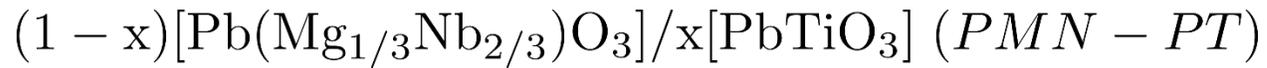
- **Reduce T1!**
- **Excite electrons!**

# DNP

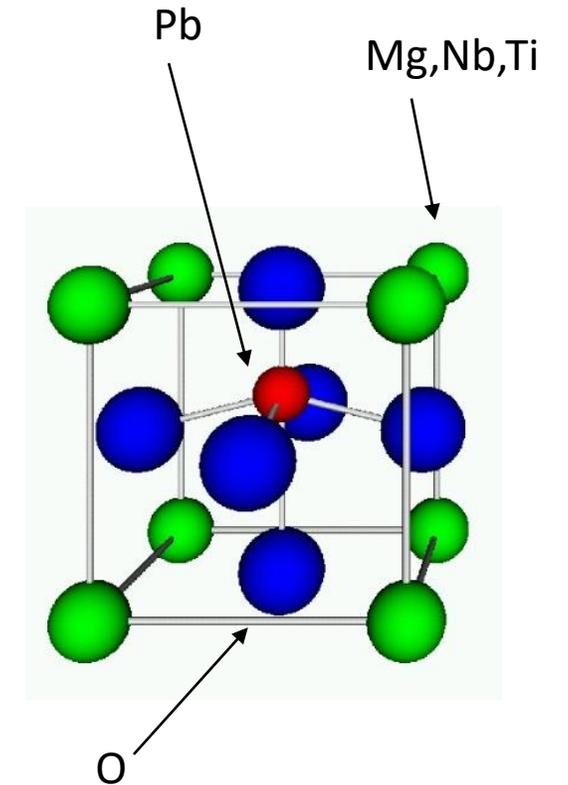
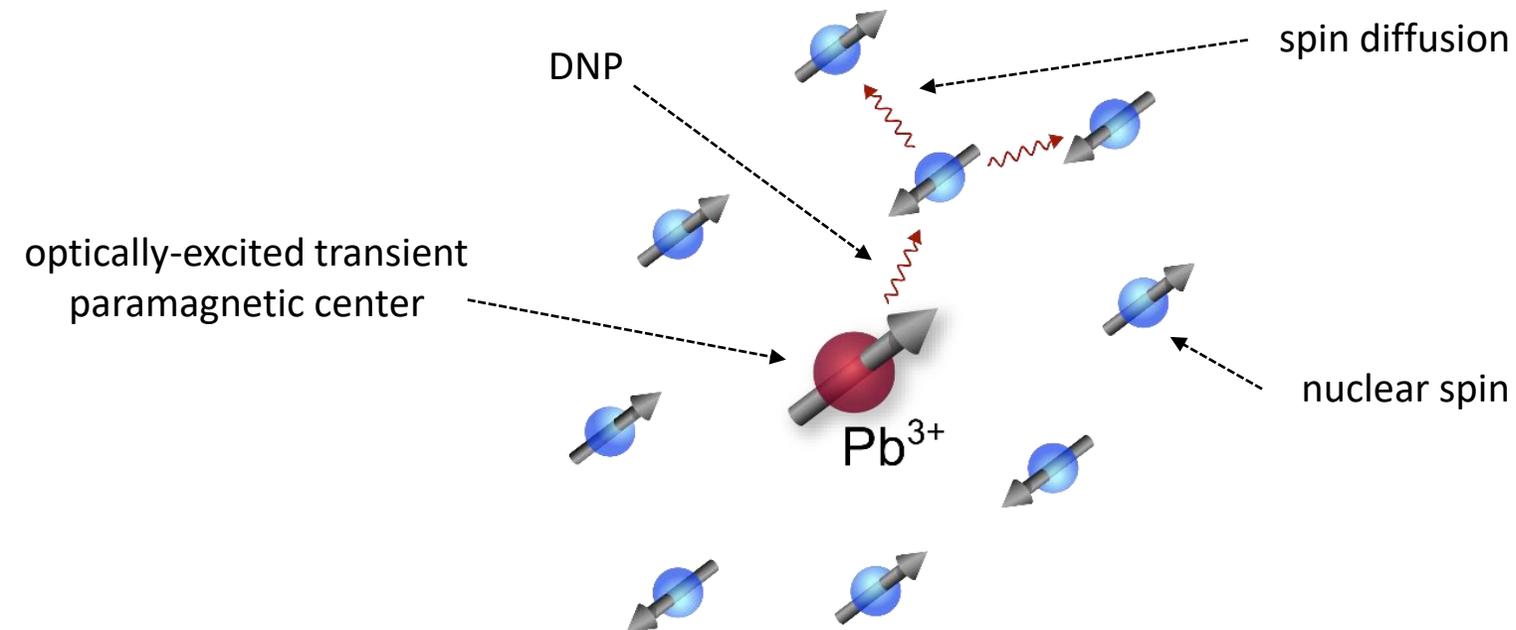
**Method of increasing polarization of nuclei above the thermal limit**

Transmit polarization from the electrons to the nuclei

Our crystal: PMN-PT



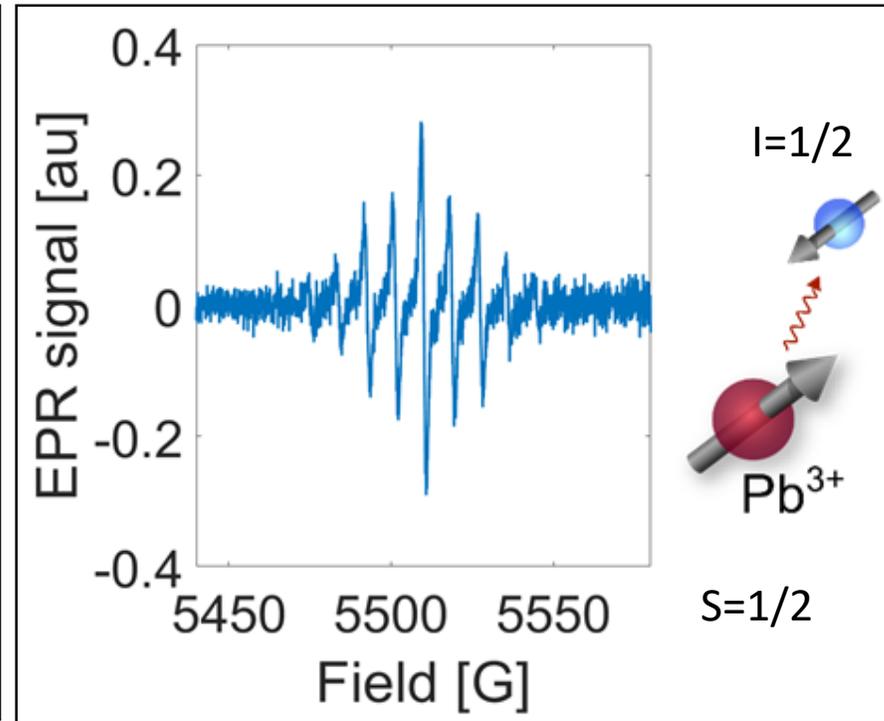
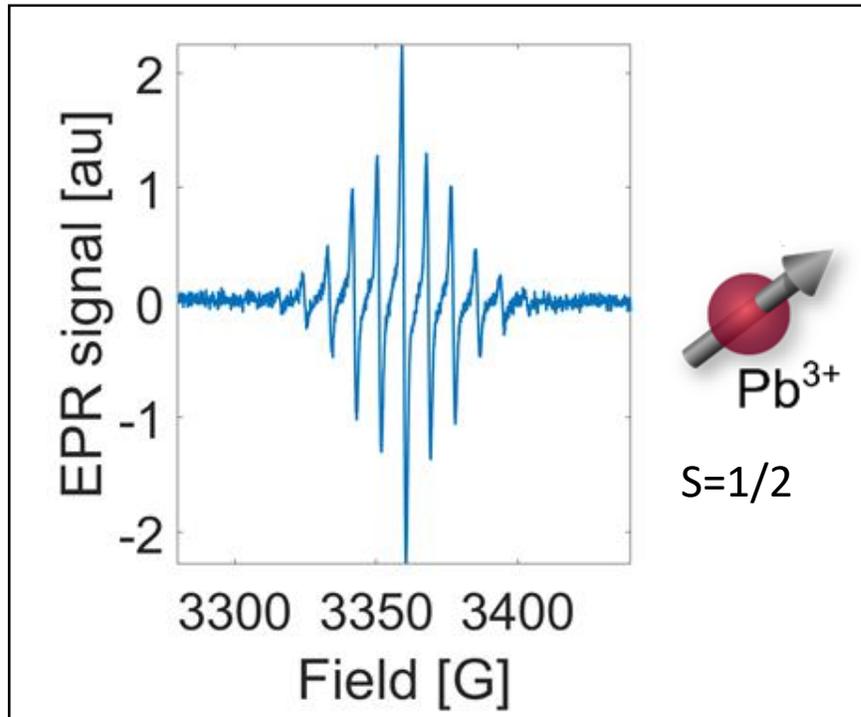
Electrons excited by 405 nm laser



# EPR basics and Lead Titanate (PT)

**EPR is similar to NMR but with electronic spins – spins are excited by microwaves**

We use a Bruker EPR with an Oxford Instruments helium cryostat - operate as low as 4 K  
Couple light using a glass rod



$I=0$ : 79% abundance ( $^{204}Pb$ ,  $^{206}Pb$ ,  $^{208}Pb$ )

$I=1/2$ : 21% abundance ( $^{207}Pb$ )

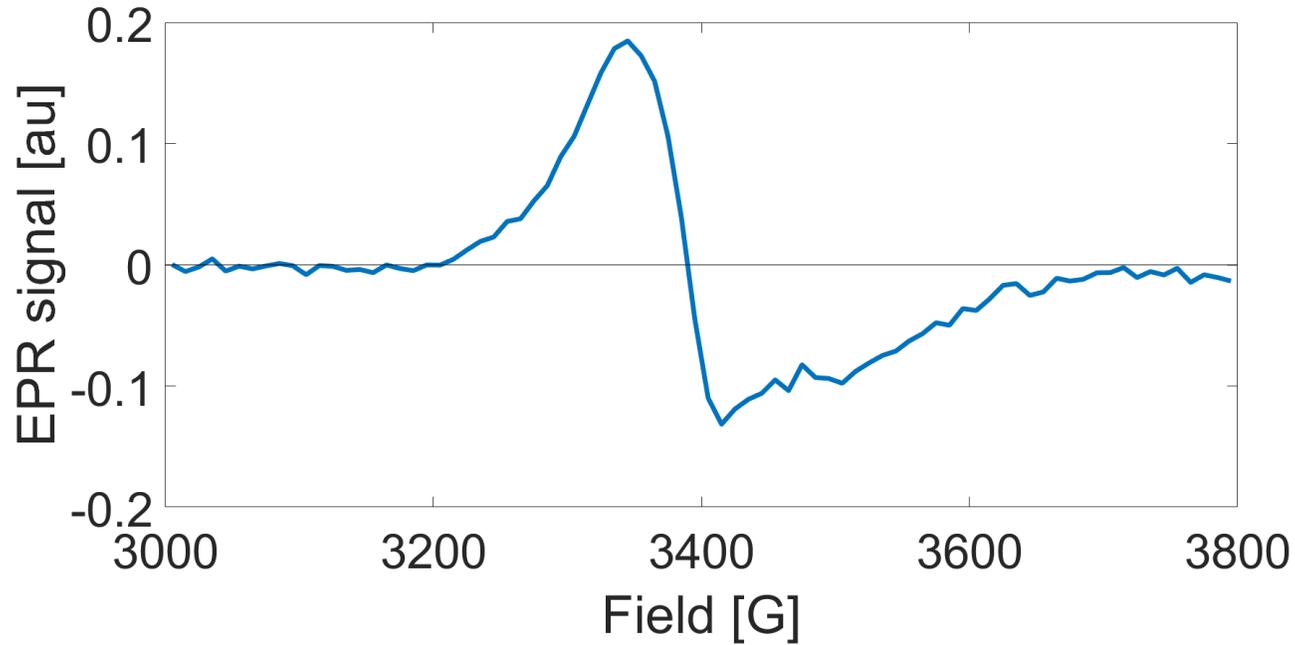
[Phys. Rev. B 53, 3080 (1996)]

Unfortunately, it is difficult to grow a large enough crystal

**Hyperfine structure proves that we excited  $Pb^{3+}$  paramagnetic centers**

# EPR results – PMN-PT

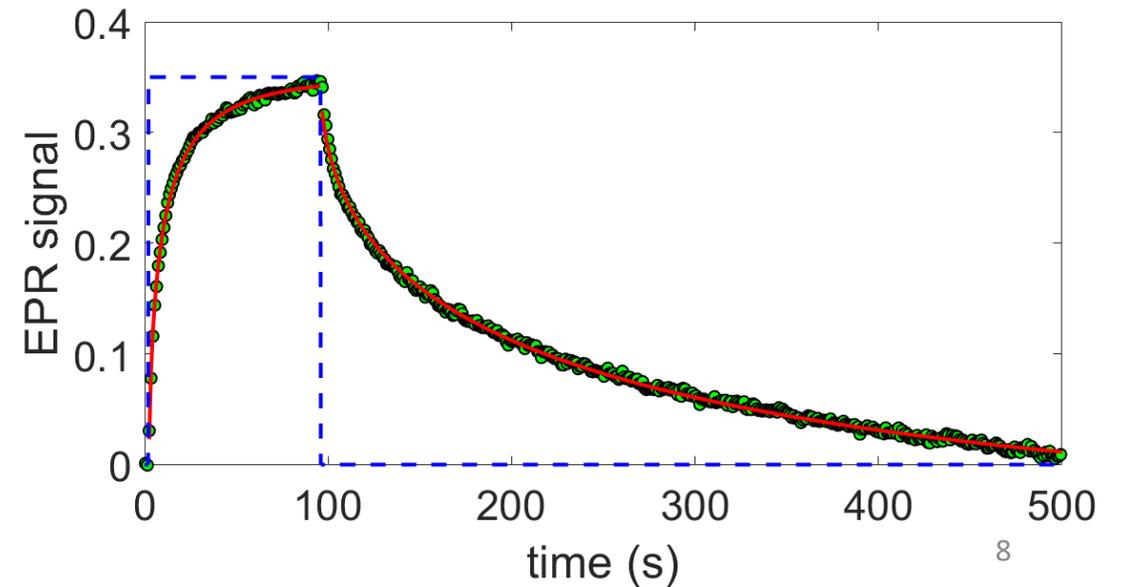
PMN-PT EPR transition at g=2 at 10K and 9.4GHz and 0.5 mW laser



Estimate number of spins by comparing to a sample with known number of spins:

$$n \sim 10^{18} \text{ cm}^{-3}$$

PMN-PT EPR decay at fixed field (3350G) at 10K, 9.4GHz and 0.5mW laser



**Excited electrons have a long lifetime**

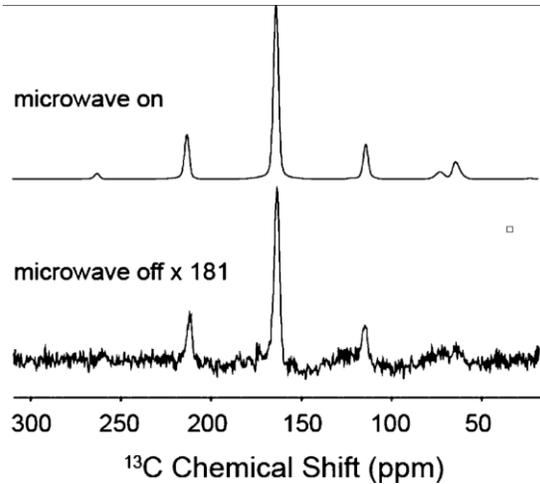
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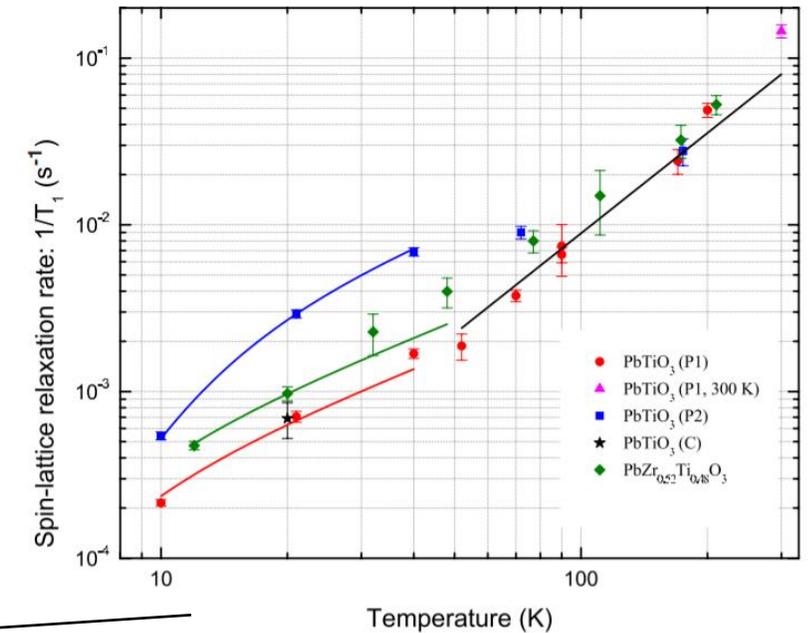
- Reduce T1!
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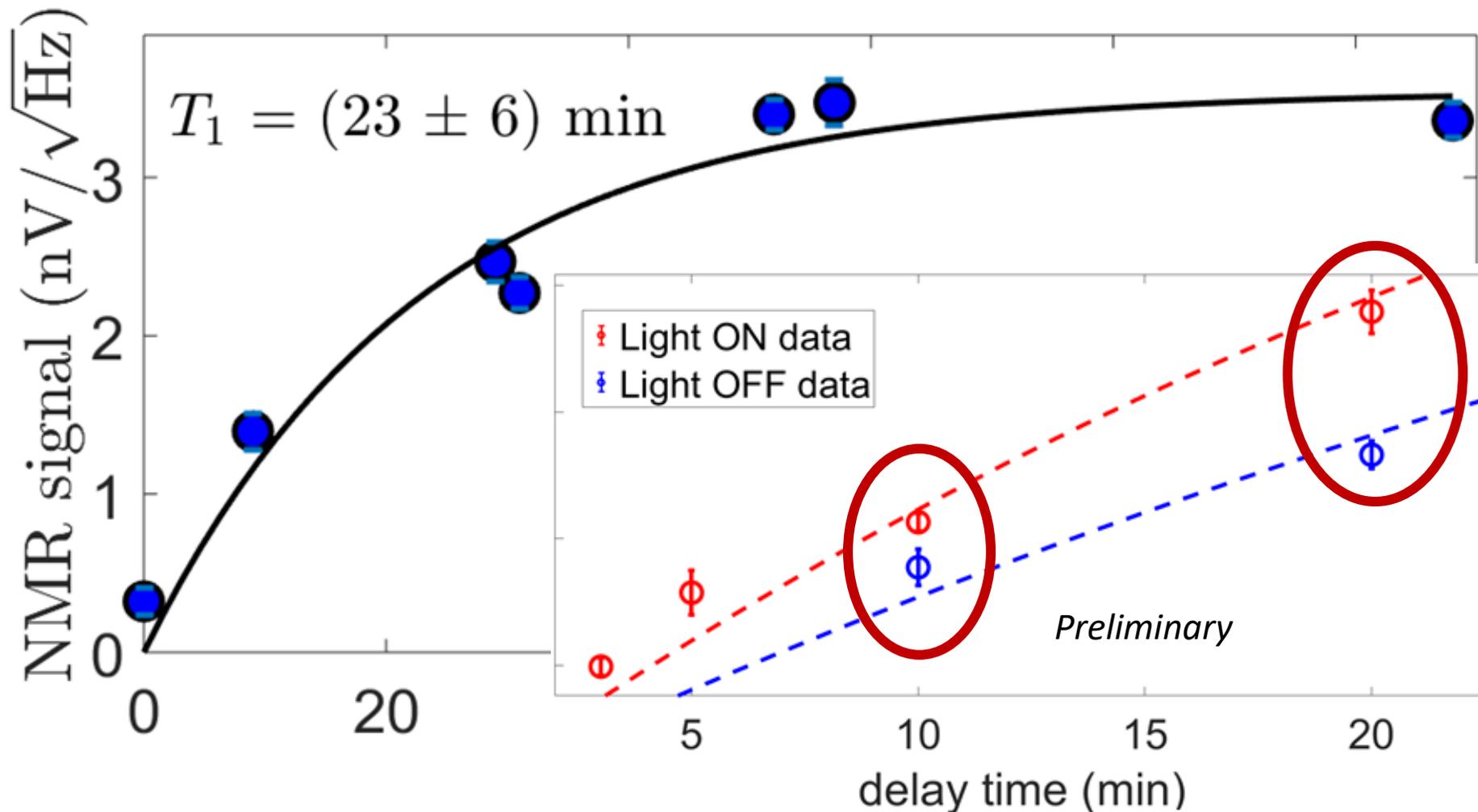


[Phys. Rev. A 77, 022102 (2008)]

# T1 in CASPEr

Measure T1 by saturation-recovery:

saturate spins by many NMR pulses and probe after a given waiting time

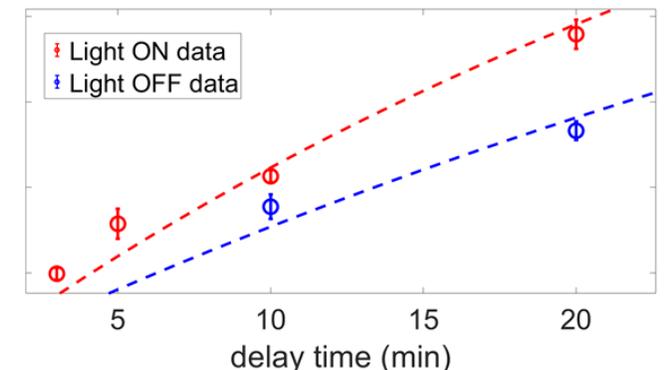
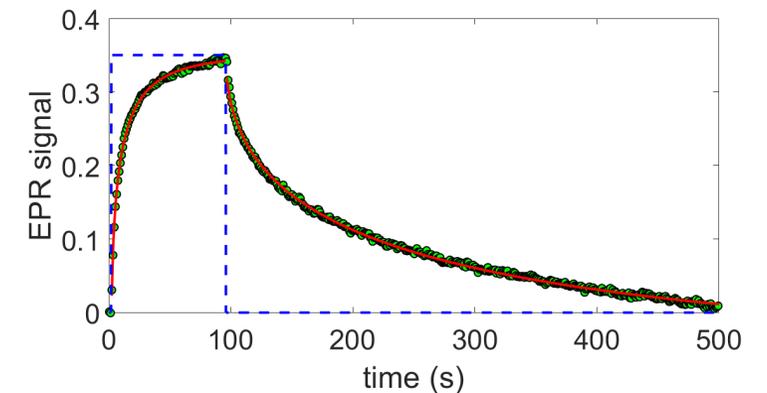
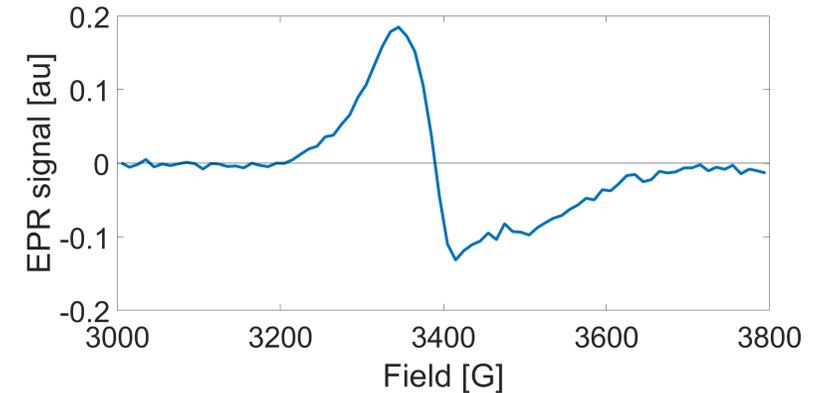


Optically excited electrons possibly reduce T1

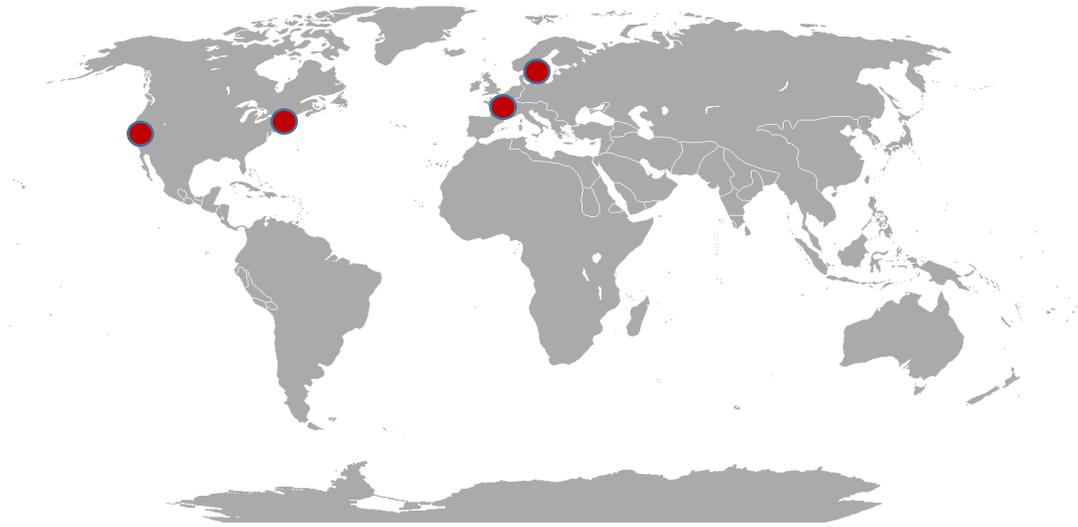
We can polarize sample faster!

# Summary and next steps

- In order to extend the experiment we can either
  - **Enhance polarization by DNP**
  - **Increase thermal polarization**
- **Illumination excites paramagnetic centers**
- First steps towards DNP:
  - **Estimated the number of electrons**
  - **Understood the lifetime of excited states**
  - Next step: understand lineshape, and decide what DNP method to implement
- We measured T1 with and without laser illumination
  - **Final polarization is restored quicker with laser**
  - Next step: quantify T1 and understand laser power dependence



## CASPER collaboration



Alfred P. Sloan  
FOUNDATION

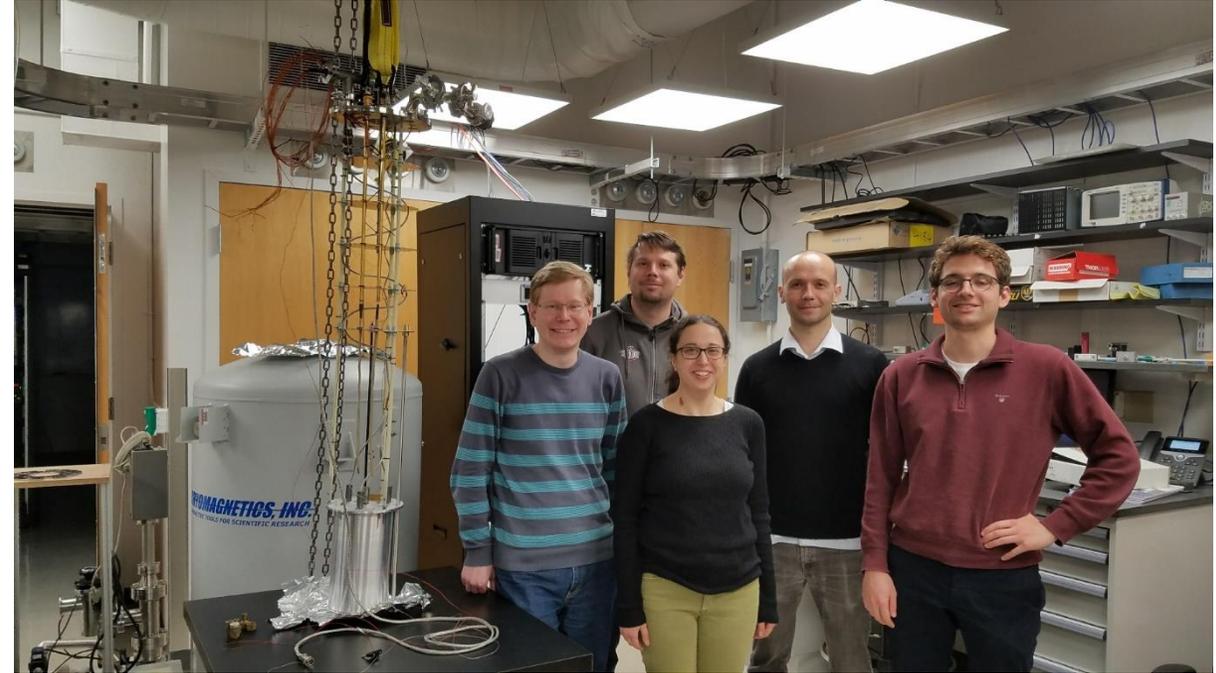
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## CASPER-Electric Team



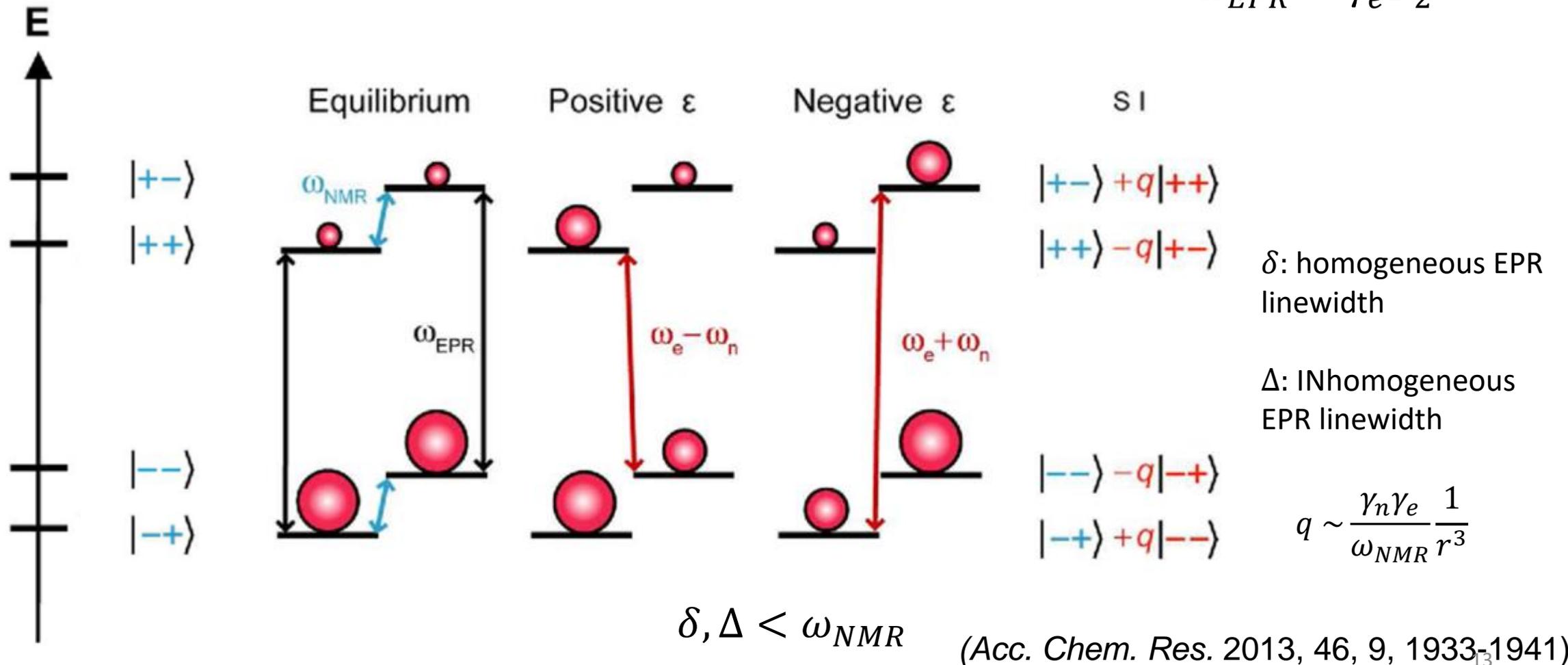
**Thank you!**

# Solid effect

$$H = \gamma_e \mathbf{S} \cdot \mathbf{B} - \gamma_n \mathbf{I} \cdot \mathbf{B} + A \mathbf{S} \cdot \mathbf{I} = \gamma_e S_z B_z - \gamma_n I_z B_z + C(I^+ S_z + I^- S_z)$$

$$\omega_{NMR} = \gamma_n B_1$$

$$\omega_{EPR} = \gamma_e B_2$$



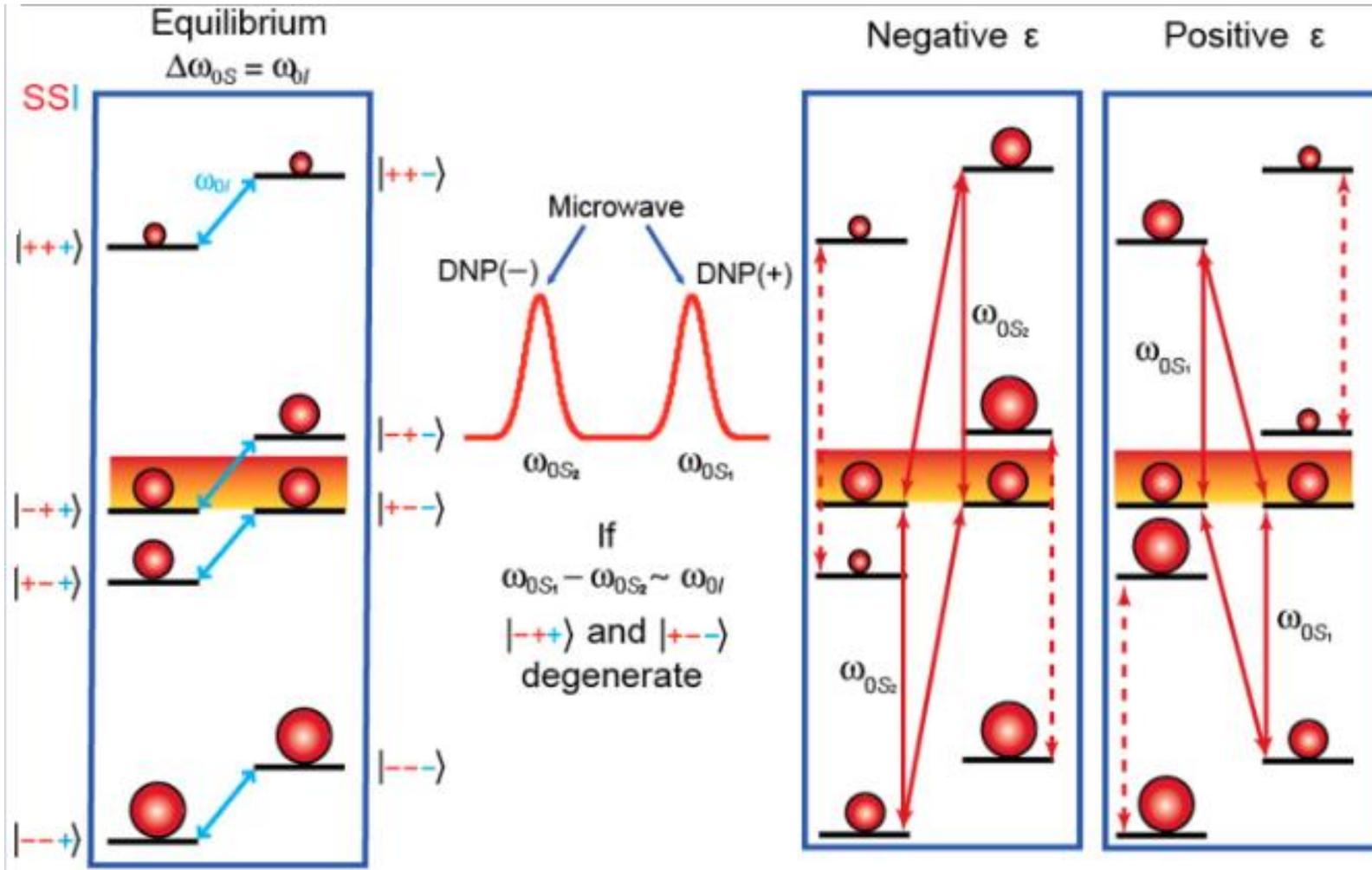
# Cross effect

$$H = \gamma_e \mathbf{S}_1 \mathbf{B} + \gamma_e \mathbf{S}_2 \mathbf{B} - \gamma_n \mathbf{I} \mathbf{B} + A \mathbf{S}_1 \mathbf{I} + D \mathbf{S}_1 \mathbf{S}_2$$

$$\omega_{01} = \gamma_n B_n$$

$$\omega_{s1} = \gamma_e B_{e1}$$

$$\omega_{s2} = \gamma_e B_{e2}$$



$\delta$ : homogeneous EPR linewidth

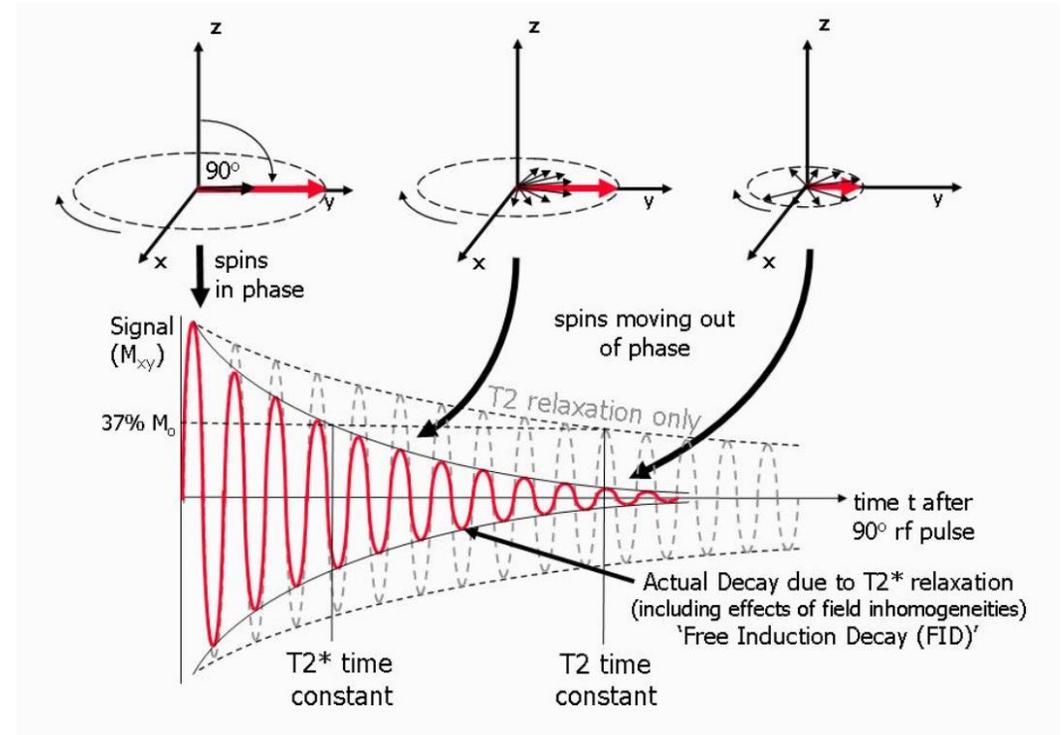
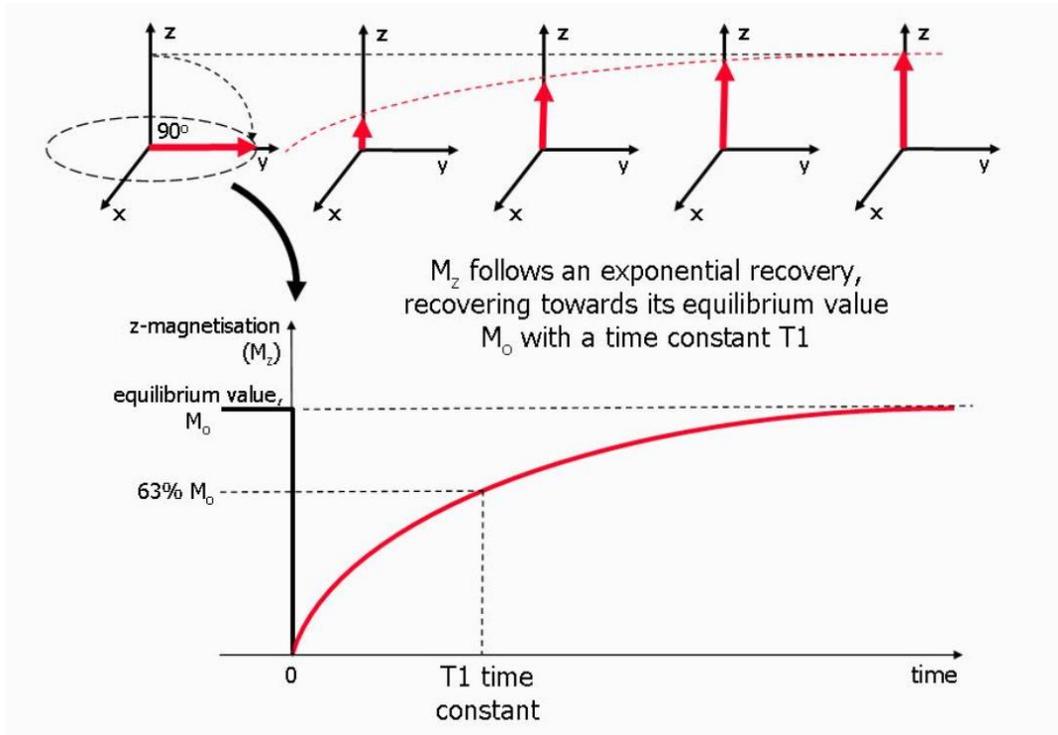
$\Delta$ : inhomogeneous EPR linewidth

$$\Delta > \omega_{NMR} > \delta \quad (\text{Acc. Chem. Res. 2013, 46, 9, 1933-1941})$$

# Relaxation times in NMR

T1 - **restore longitudinal** magnetization – also called spin-lattice relaxation

T2 - **lose transversal** magnetization – also called spin-spin relaxation



## Advantages and disadvantages of a long T1

- Able to pre polarize spins at high magnetic field and then sweep to lower fields, preserving large polarization
- Subsequent NMR pulses saturate spins and can't do NMR until spins are not recovered