

# Circuit-QED-enhanced magnetic resonance

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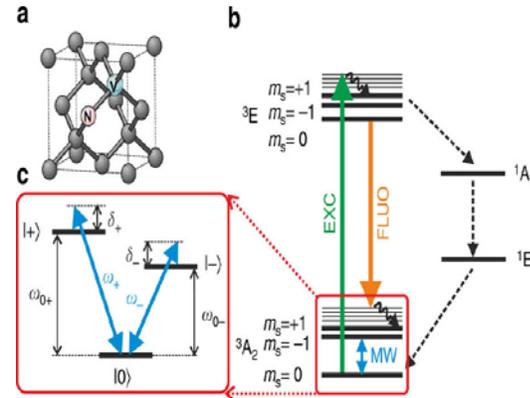
G. Zhang  
R. Liu



# Nanoscale magnetic resonance

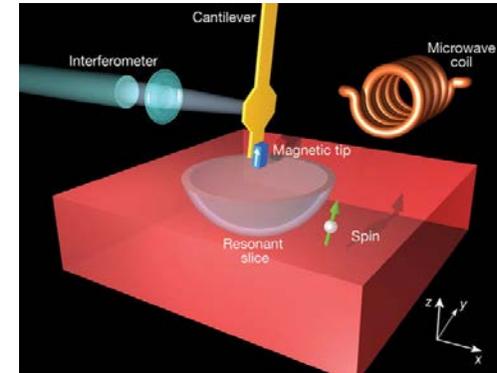


Fantastically useful but  
 Low sensitivity  
 Macroscopic samples



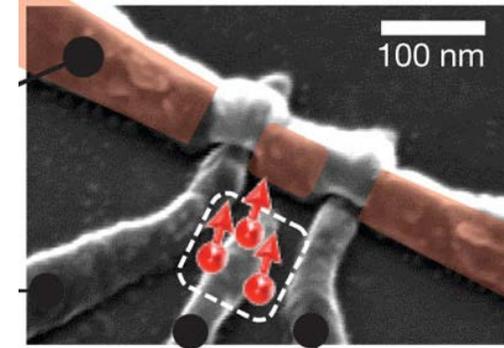
## Optical detection

Nature **363**, 244 (1993)  
 Nat. Com. **5**, 4870 (2014)  
 Nat. Nano. **9**, 279 (2014)



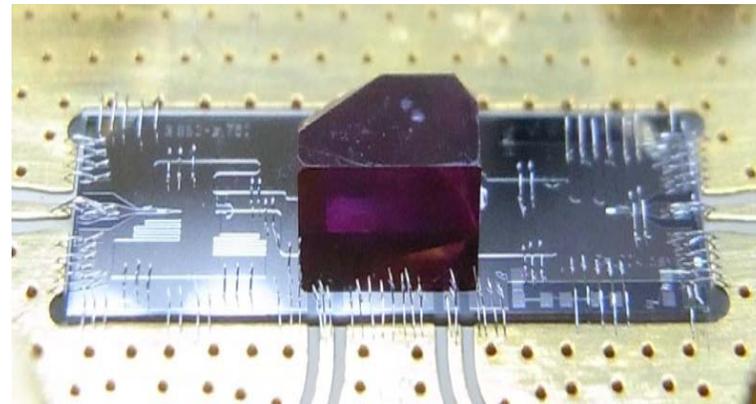
## Scanning probe

Science **350**, 417 (2015)  
 Nature **430**, 329 (2004)  
 Phys. Rev. Lett. **62**, 2531 (1989)



## Electrical detection

Rev. Sci. Instr. **83**, 043907 (2012)  
 Nature **467**, 687 (2010)



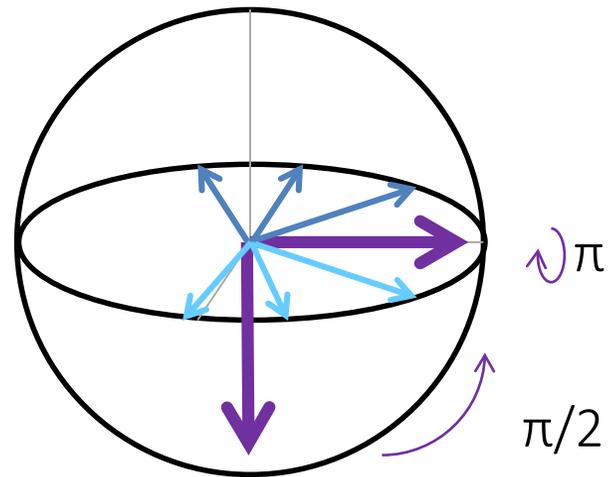
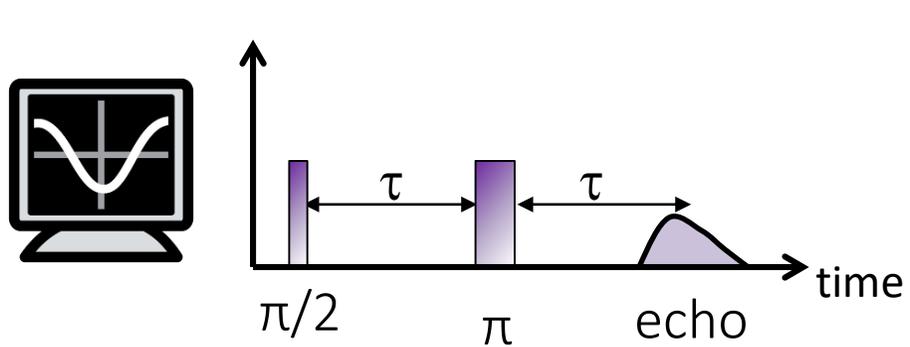
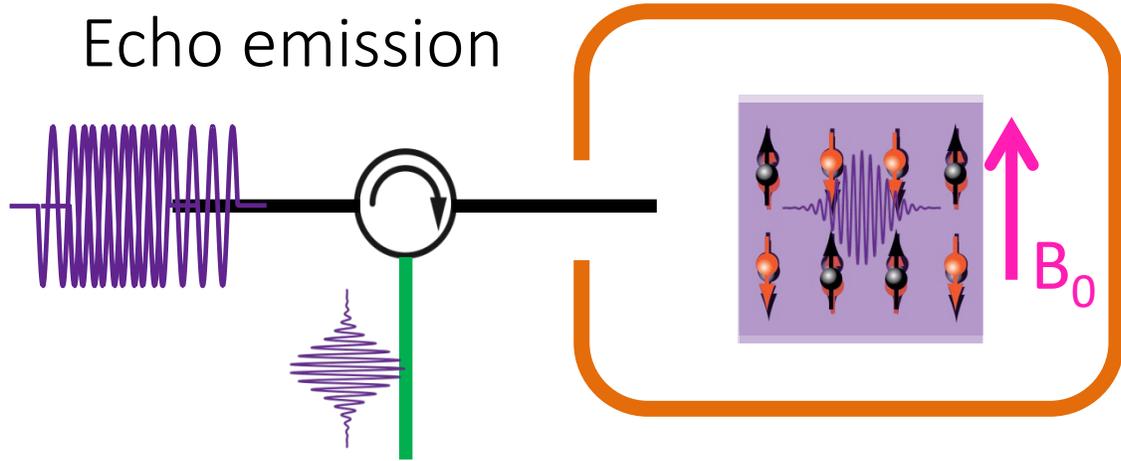
## Circuit QED for electron spin resonance detection

- 1) Principle
- 2) Applications :
  - Few-nuclear spin detection
  - Novel e spin hyperpolarization scheme

# Conventional Pulsed Inductive Detection Electron Spin Resonance (ESR)

Excite spins

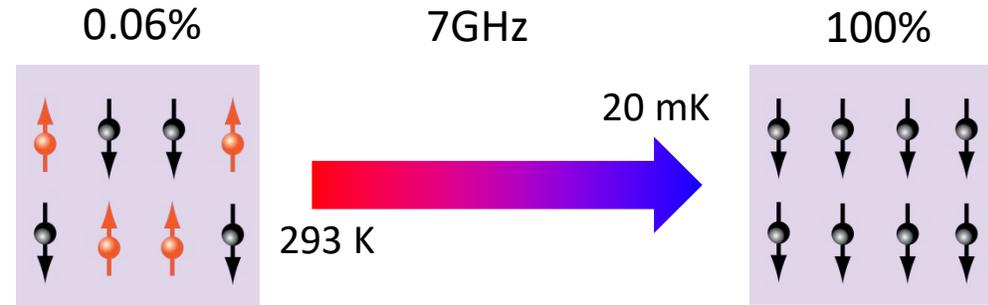
Echo emission



# Circuit QED-enhanced ESR

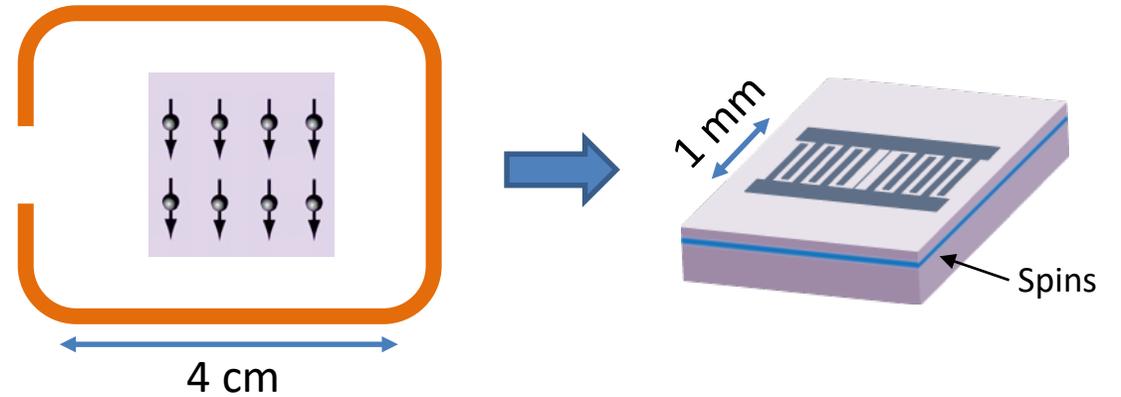
## 1. Low Temperature

- *Maximum spin Polarization*
- *No thermal noise*



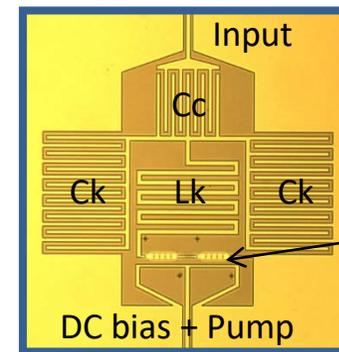
## 2. Superconducting Micro-Resonators

- *Large spin-MW photon coupling  $g$*
- *Large quality factor  $Q$*



## 3. Quantum limited detection chain

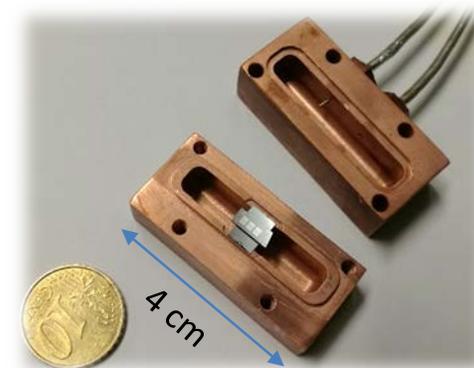
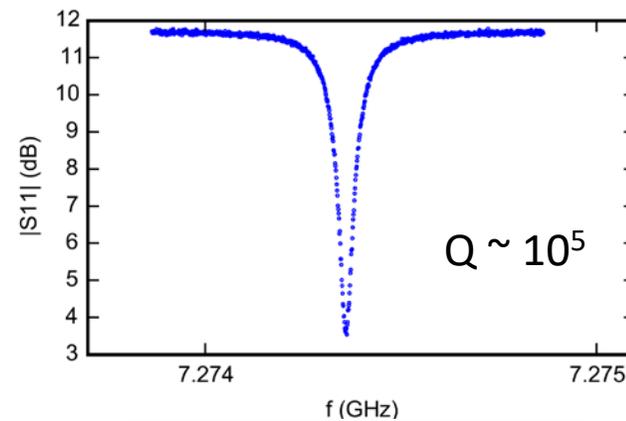
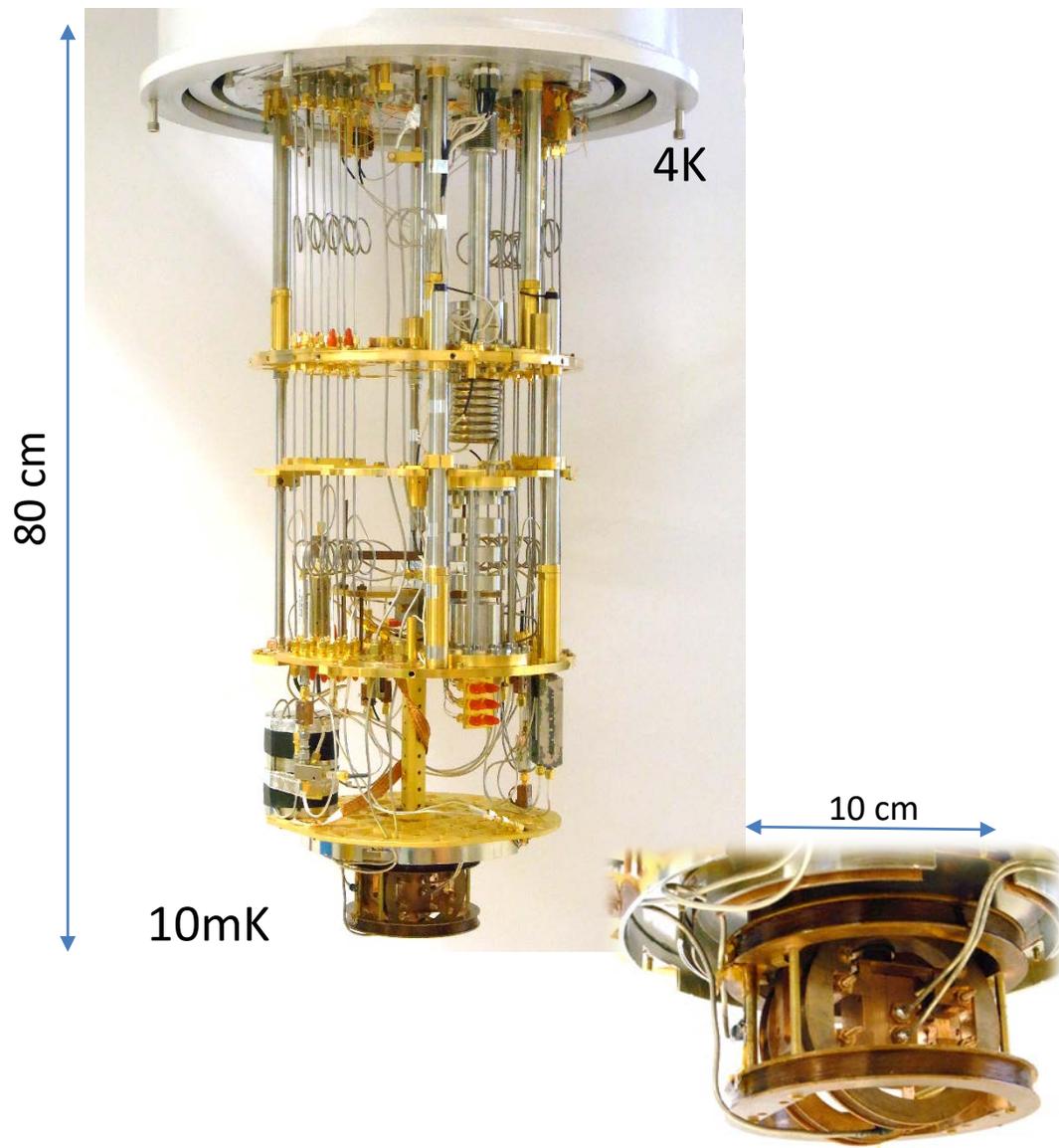
- *"Noiseless" amplification by superconducting Josephson Parametric Amplifier (JPA)*



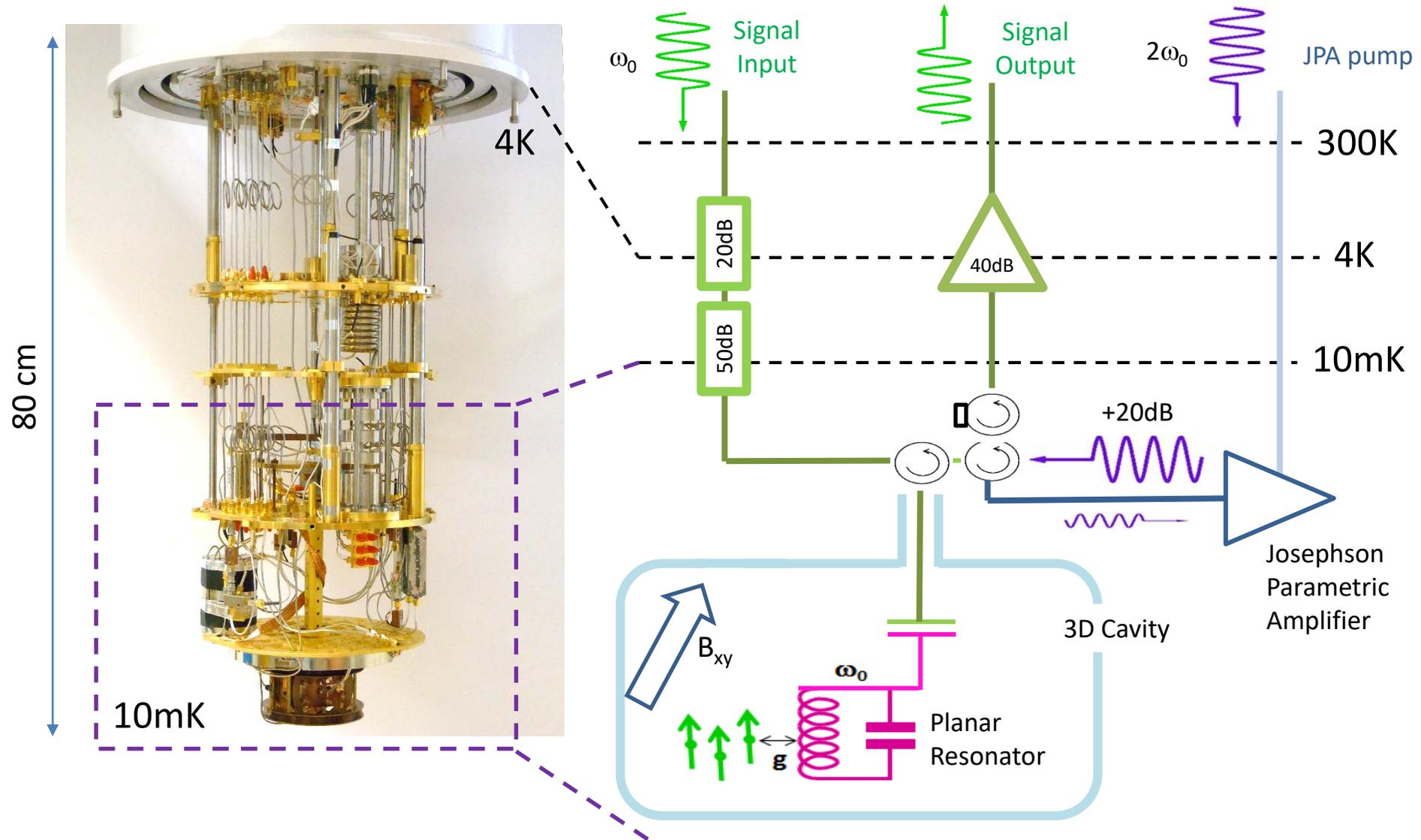
squid array

X. Zhou et al.,  
Phys. Rev. B **89**, 214517 (2014)

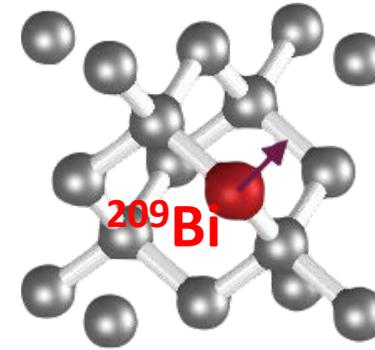
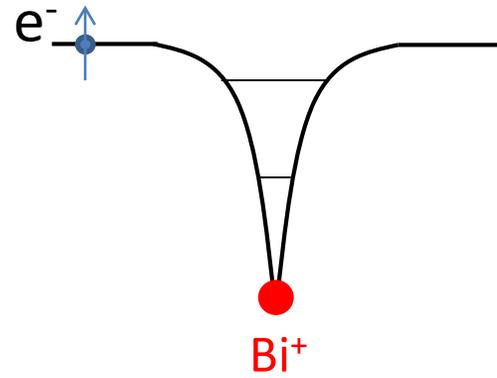
# Circuit QED-enhanced ESR : setup



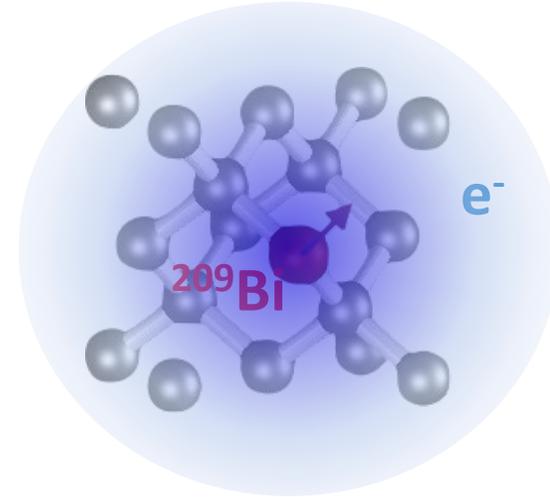
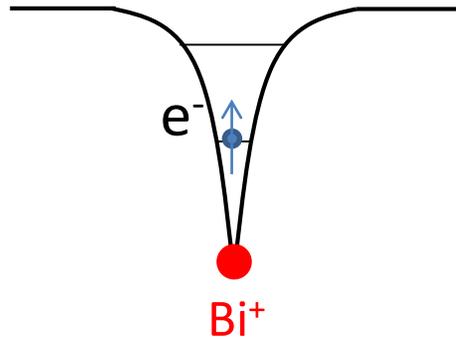
# Circuit QED-enhanced ESR : setup



# Bismuth donors in silicon



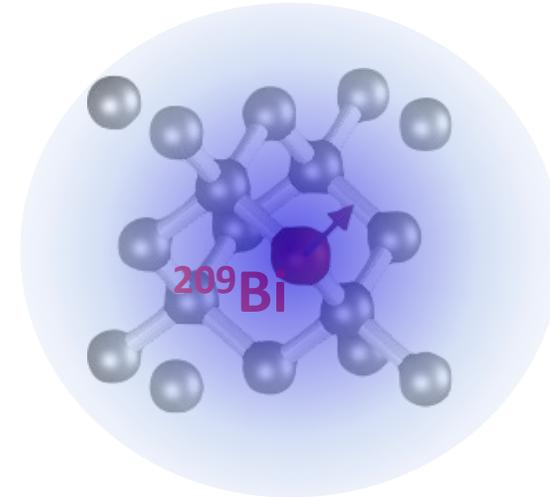
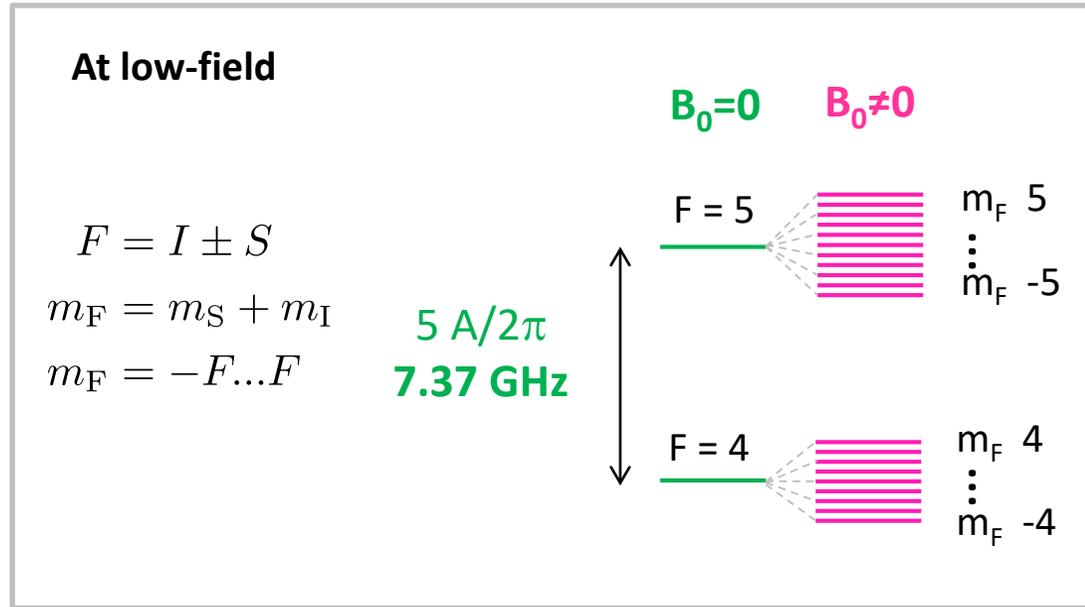
# Bismuth donors in silicon



$$\frac{H}{\hbar} = \underbrace{AI \cdot S}_{\text{HYPERFINE}} + \underbrace{B_0 \cdot (-\gamma_e S - \gamma_n I)}_{\text{ZEEMAN EFFECT}}$$

- Electronic spin = 1/2
  - Nuclear spin I=9/2
  - Large hyperfine coupling  $\frac{A}{2\pi} = 1.4754\text{GHz}$
- } 20 electro-nuclear states

# Bismuth donors in silicon

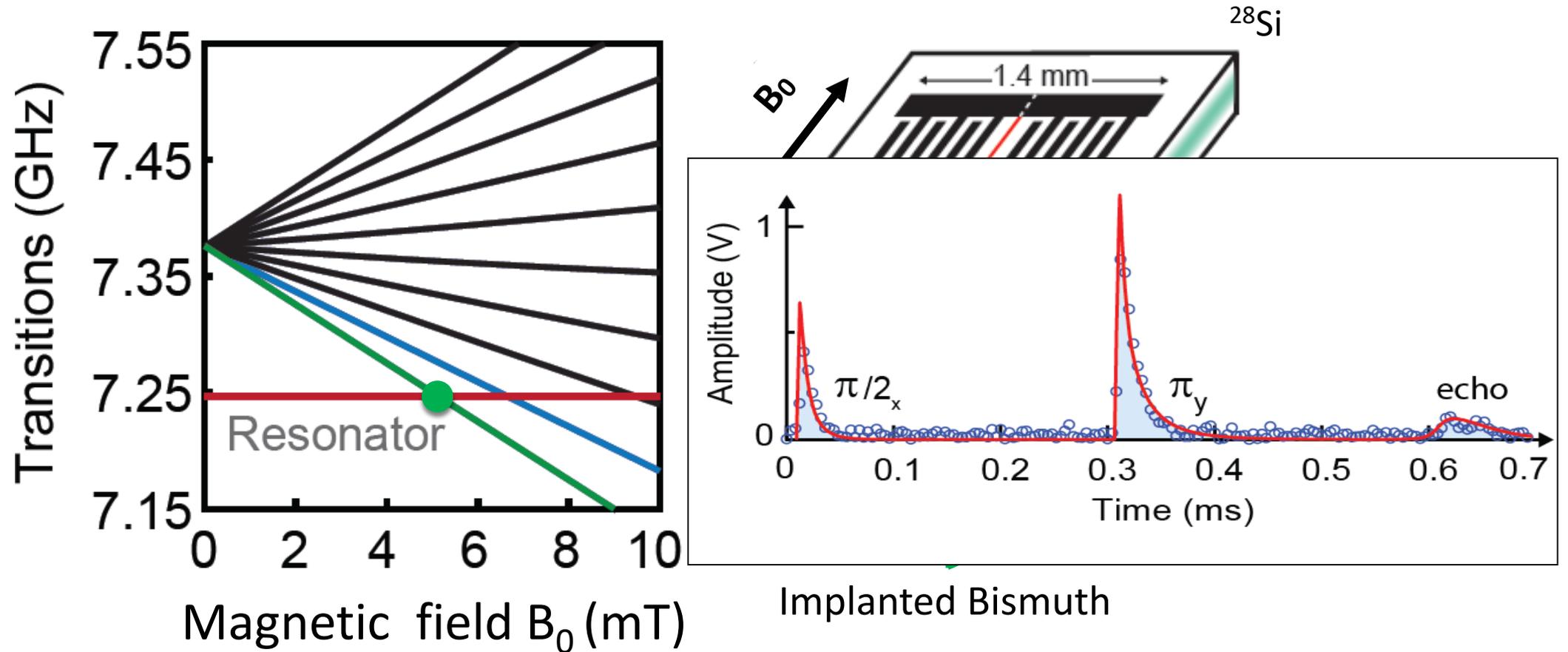


$$\frac{H}{\hbar} = \underbrace{AI \cdot S}_{\text{HYPERFINE}} + \underbrace{B_0 \cdot (-\gamma_e S - \gamma_n I)}_{\text{ZEEMAN EFFECT}}$$

- Nuclear spin  $I=9/2$
- Electronic spin  $S=1/2$
- Large hyperfine coupling  $\frac{A}{2\pi} = 1.48 \text{ GHz}$

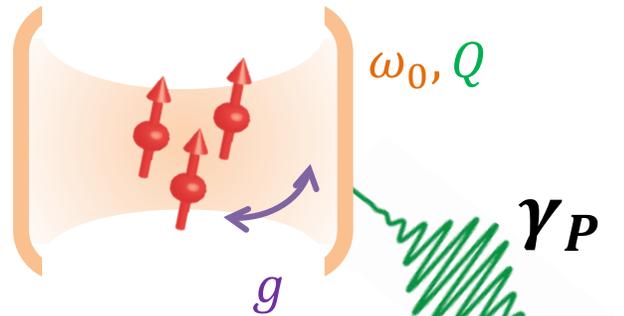
# Bismuth donors in silicon

10 allowed ESR-like transitions @ low  $B_0$



•  $m_F = 4 \rightarrow m_F = 5$ , @ ~5 mT

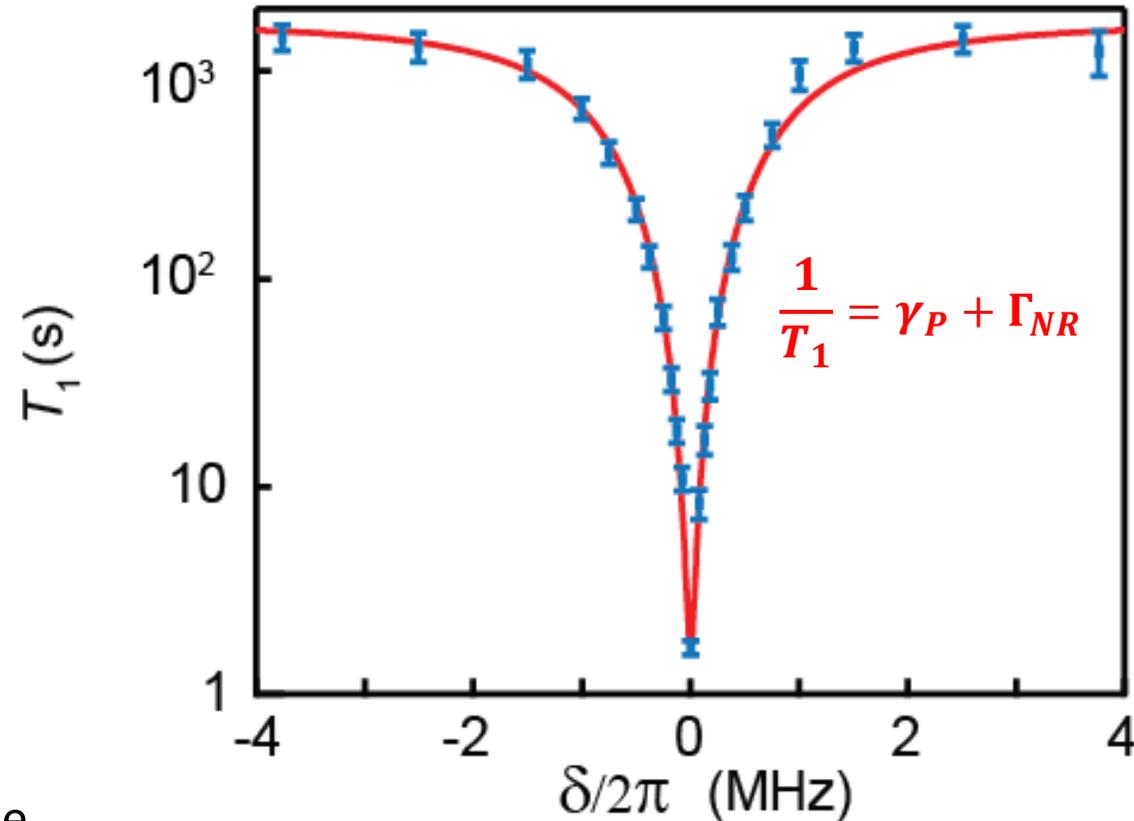
# Spin relaxation by spontaneous emission through the cavity



$$\gamma_P = \frac{4Qg^2}{\omega_0} \frac{1}{1 + 4Q^2 \left[ \frac{\omega_s - \omega_0}{\omega_0} \right]^2}$$

At resonance,  $\gamma_P = 4Qg^2/\omega_0$   
Direct experimental determination of the  
spin-photon coupling constant

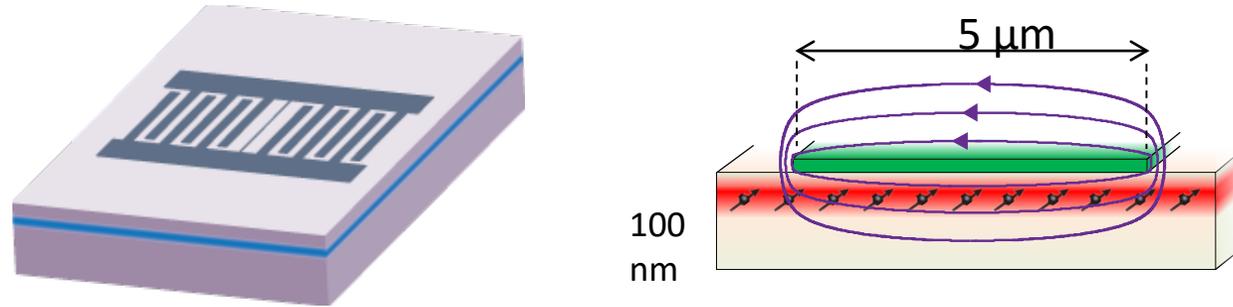
$$\frac{g}{2\pi} = 60 \pm 10 \text{ Hz}$$



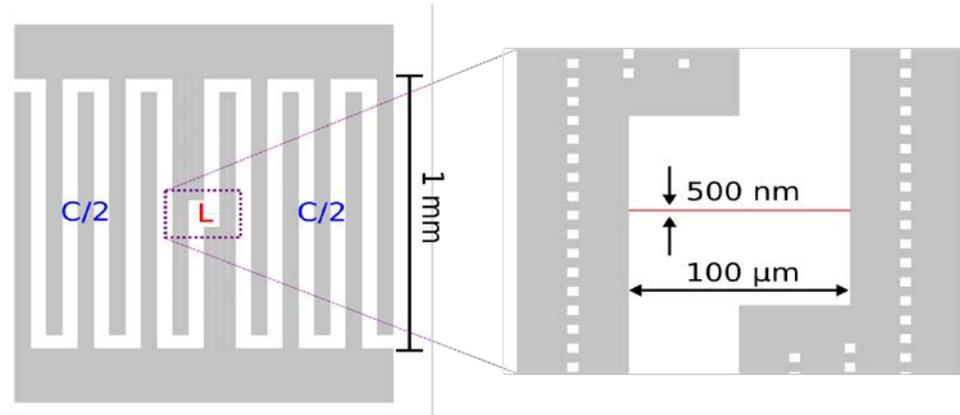
A. Bienfait et al., Nature (2016)

# Reducing the magnetic mode volume to enhance the coupling

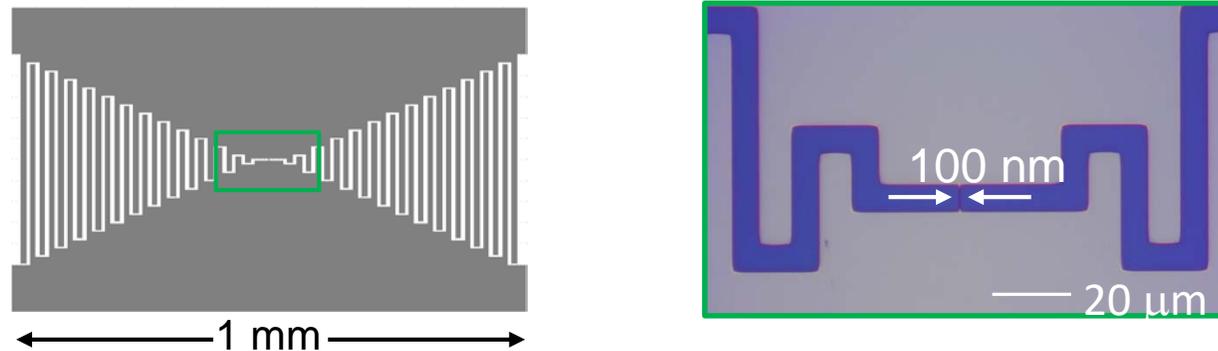
$L=1\text{mm}$   
 $w=5\mu\text{m}$   
 $V \approx 20pL$



$L=0.1\text{mm}$   
 $w=0.5\mu\text{m}$   
 $V \approx 200fL$

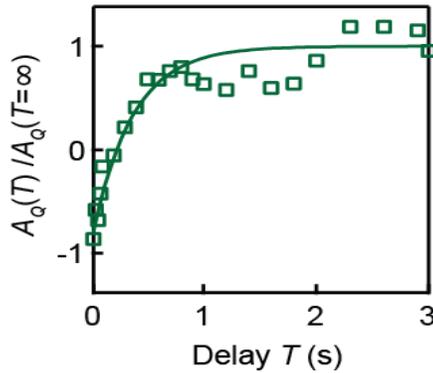


$L=10\mu\text{m}$   
 $w=0.1\mu\text{m}$   
 $V \approx fL$



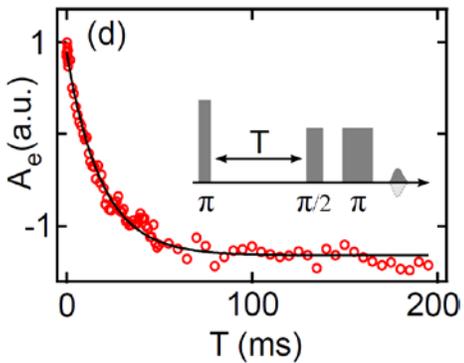
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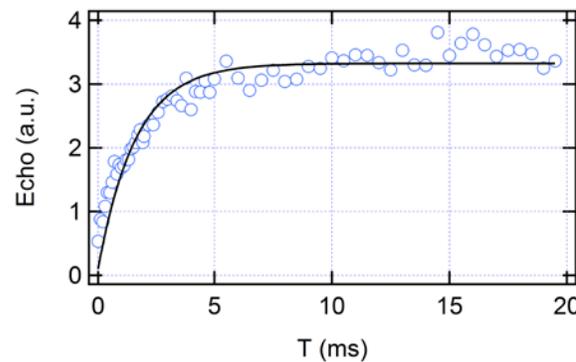
$T_1 = 0.4\text{s}$   
 $\frac{g}{2\pi} = 50\text{Hz}$   
 $2000\text{spin}/\sqrt{\text{Hz}}$

$L=0.1\text{mm}$   
 $w=0.5\mu\text{m}$   
 $V \approx 200fL$



$T_1 = 20\text{ms}$   
 $\frac{g}{2\pi} = 400\text{Hz}$   
 $65\text{spin}/\sqrt{\text{Hz}}$

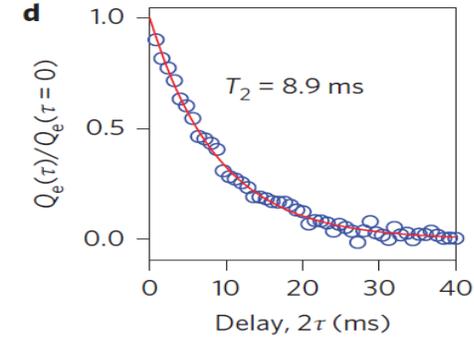
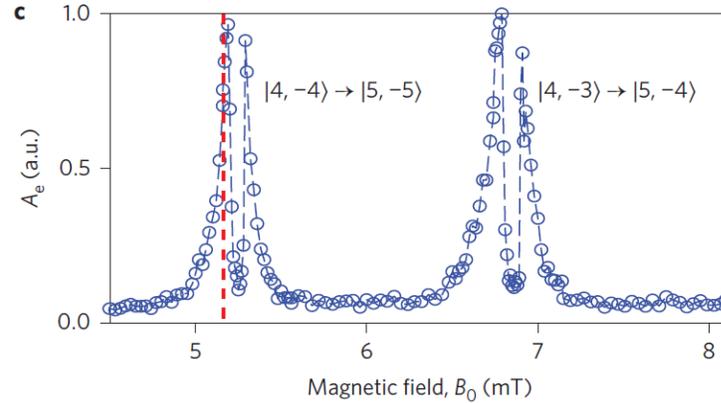
$L=10\mu\text{m}$   
 $w=0.1\mu\text{m}$   
 $V \approx fL$



$T_1 = 1.3\text{ms}$   
 $\frac{g}{2\pi} = 3\text{kHz}$   
 $10\text{spin}/\sqrt{\text{Hz}}$

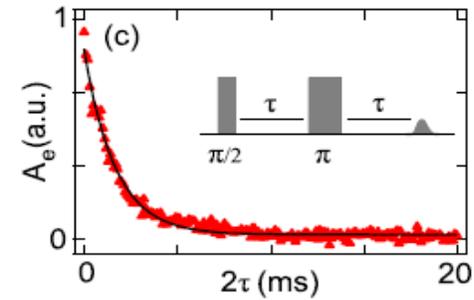
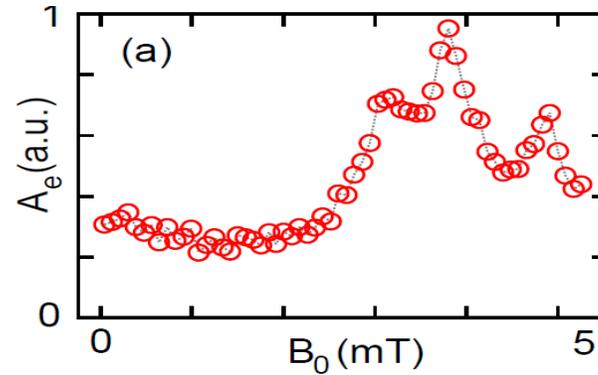
# Effect on the spin linewidth and coherence time

$L=1\text{mm}$   
 $w=5\mu\text{m}$   
 $V \approx 20pL$



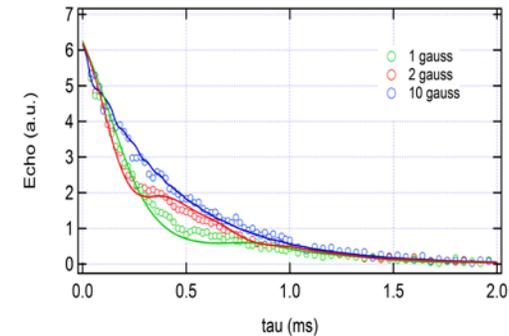
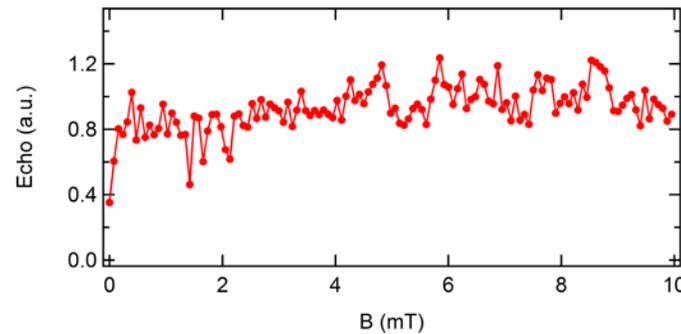
$\Gamma \approx 3\text{MHz}$   
 $T_2 = 9\text{ms}$

$L=0.1\text{mm}$   
 $w=0.5\mu\text{m}$   
 $V \approx 200fL$



$\Gamma \approx 30\text{MHz}$   
 $T_2 = 2\text{ms}$

$L=10\mu\text{m}$   
 $w=0.1\mu\text{m}$   
 $V \approx fL$

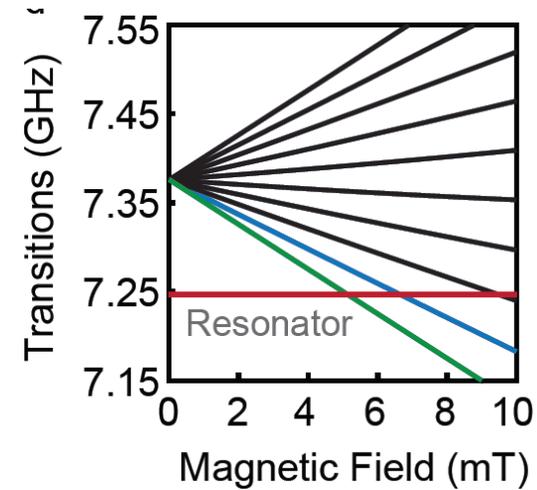


$\Gamma \approx 100\text{MHz}?$   
 $T_2 = 0.9\text{ms}$

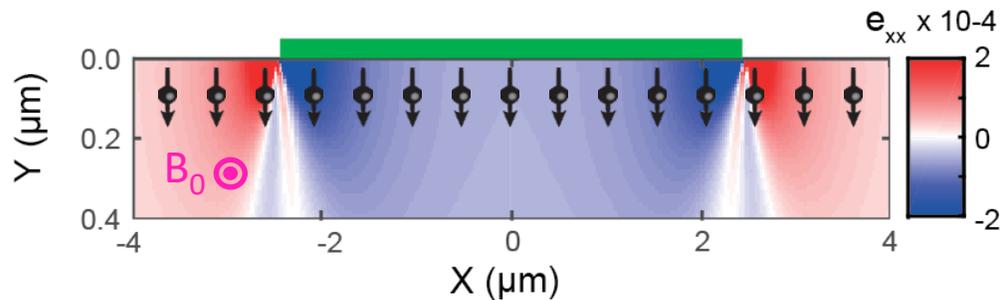
# Broadening mechanisms

$$\frac{H}{\hbar} = \mathbf{A}\mathbf{I} \cdot \mathbf{S} + \mathbf{B}_0 \cdot (-\gamma_e \mathbf{S} - \gamma_n \mathbf{I})$$

$\mathbf{A}(E, e)$  : spin line broadening if inhomogeneous electric or strain fields

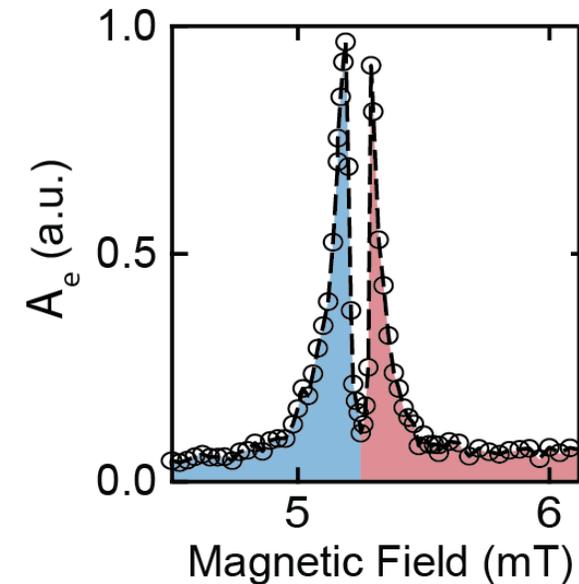


Strain induced by the aluminum wire on the underlying silicon substrate due to differential thermal contraction during cooldown

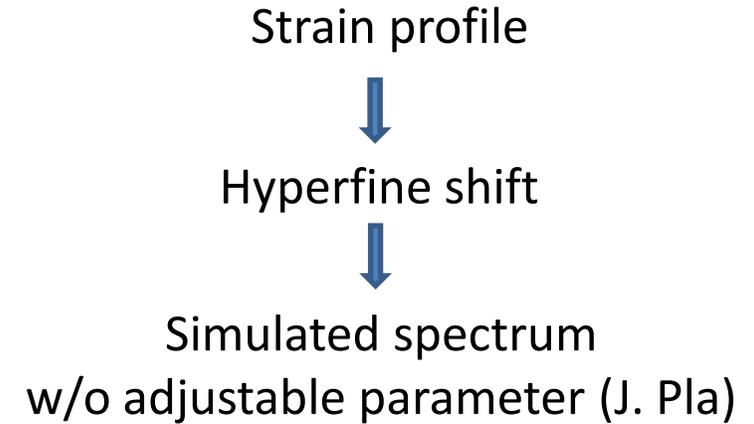
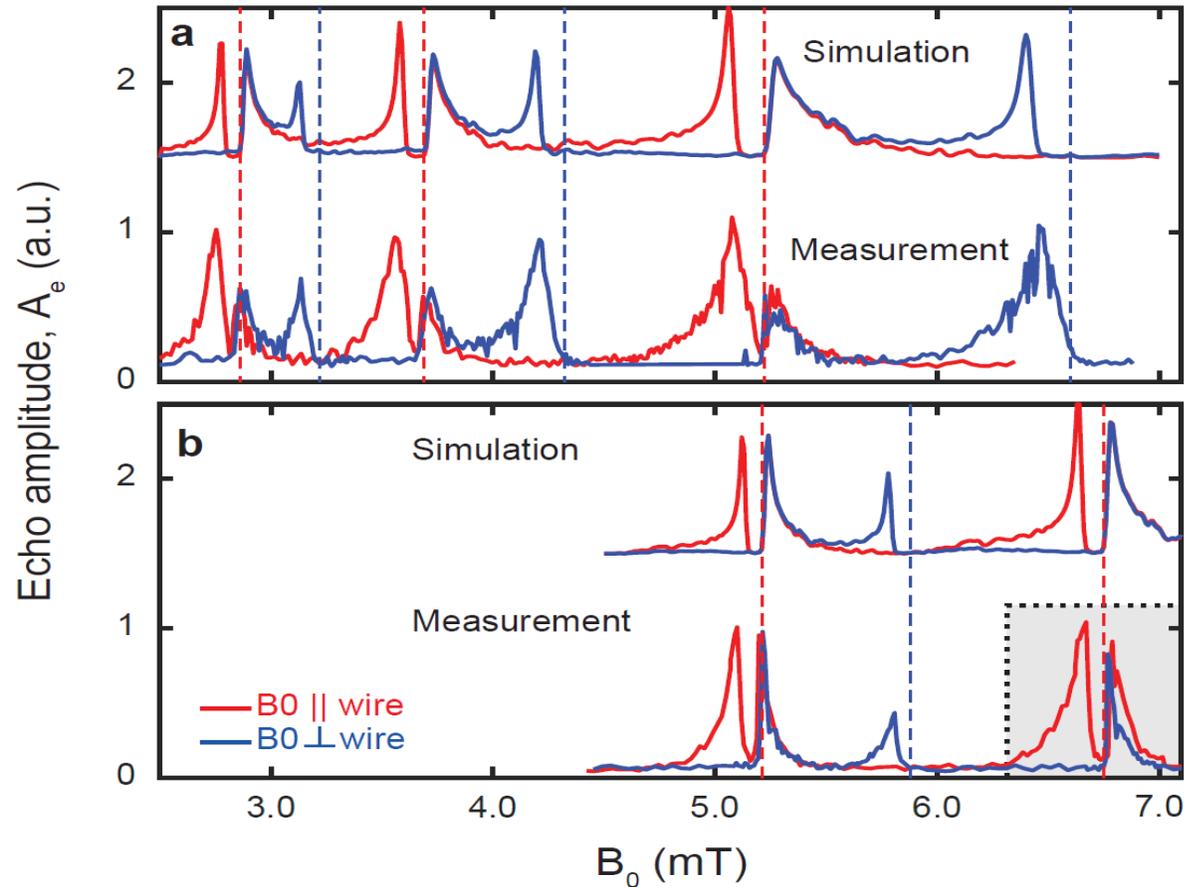


At low strain,  $A = K/3 (e_{xx} + e_{yy} + e_{zz})$  with  $K = 19\text{GHz}$

J. Mansir et al., arxiv (2017)



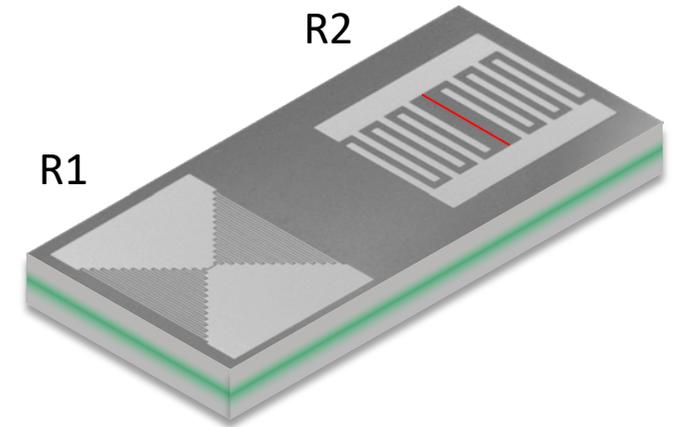
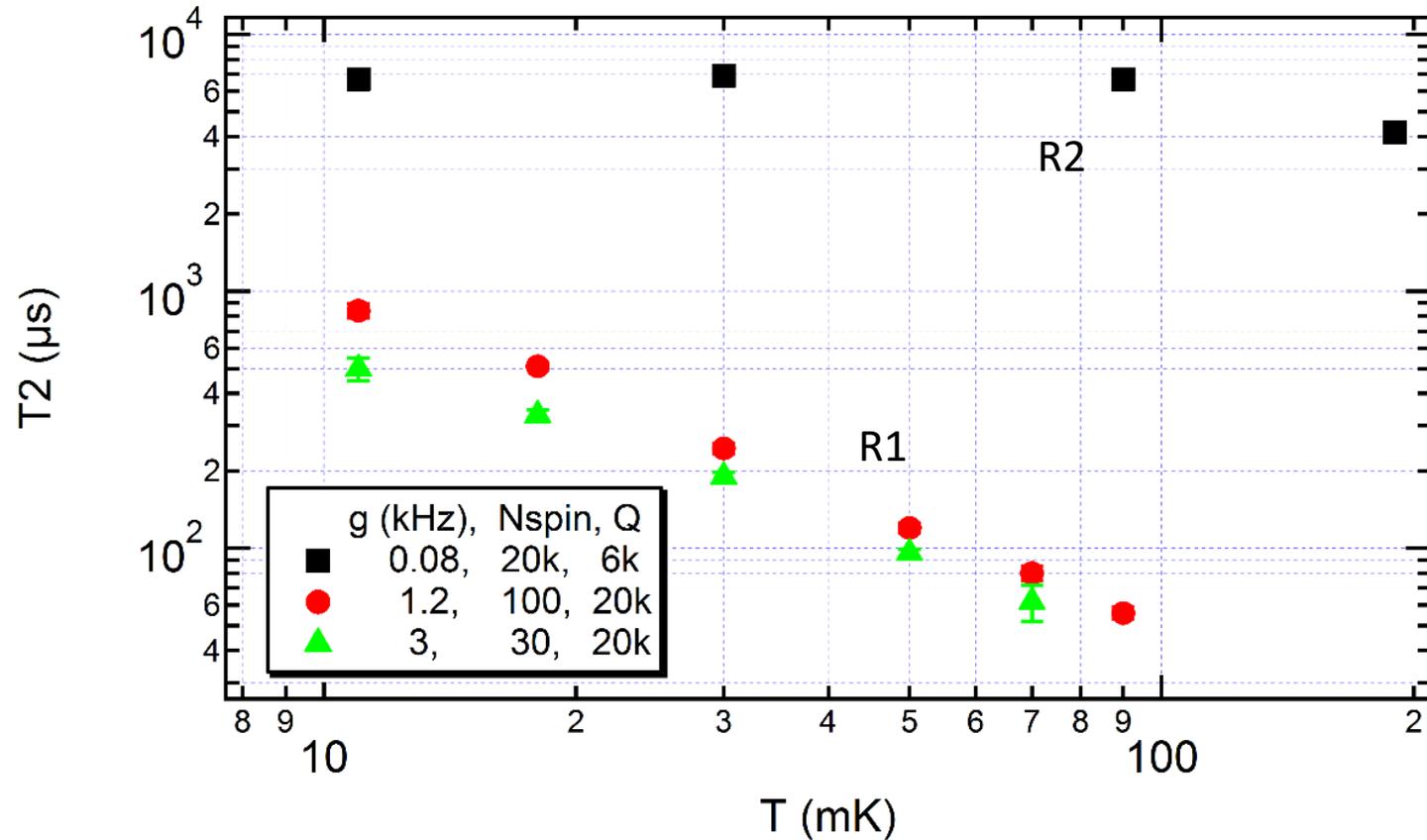
# Understanding lineshapes



J. Pla et al.,  
Phys. Rev Appl. (2018)

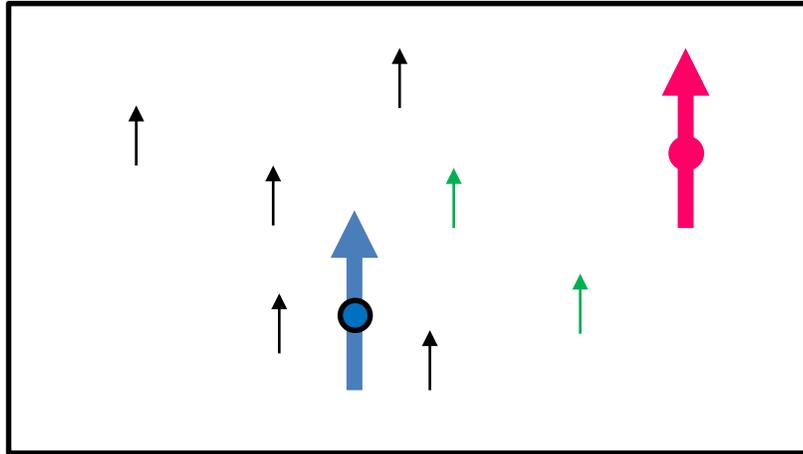
- Strain broadening : quantitative understanding for  $5\mu\text{m}$ -wide wires  
....but only qualitative for narrower wires. Extra shifts due to stray electric fields ?
- Observed reduction in  $T_2$  for increased spin-resonator coupling ??

## Temperature dependence of T2



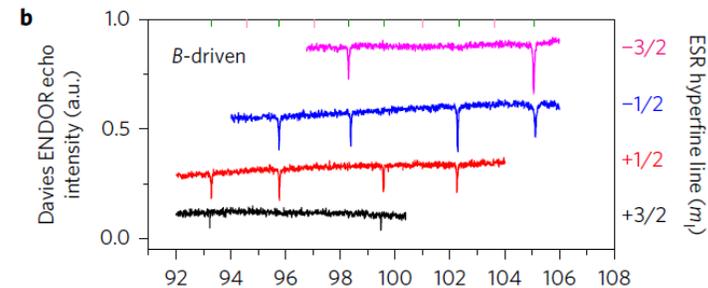
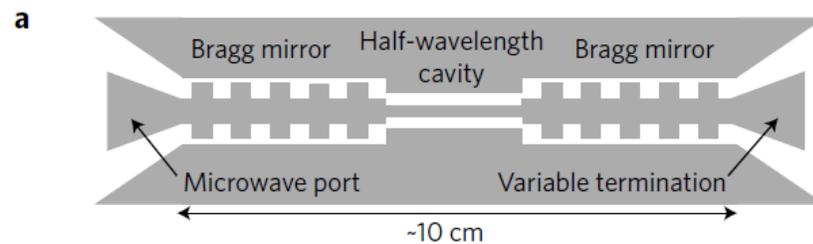
- Unexplained temperature dependence of  $T_2$ . Similar data obtained by H. Huebl's group (TUMunich)
- Non-trivial resonator-induced decoherence mechanism ?

# Accessing the electron spin magnetic environment



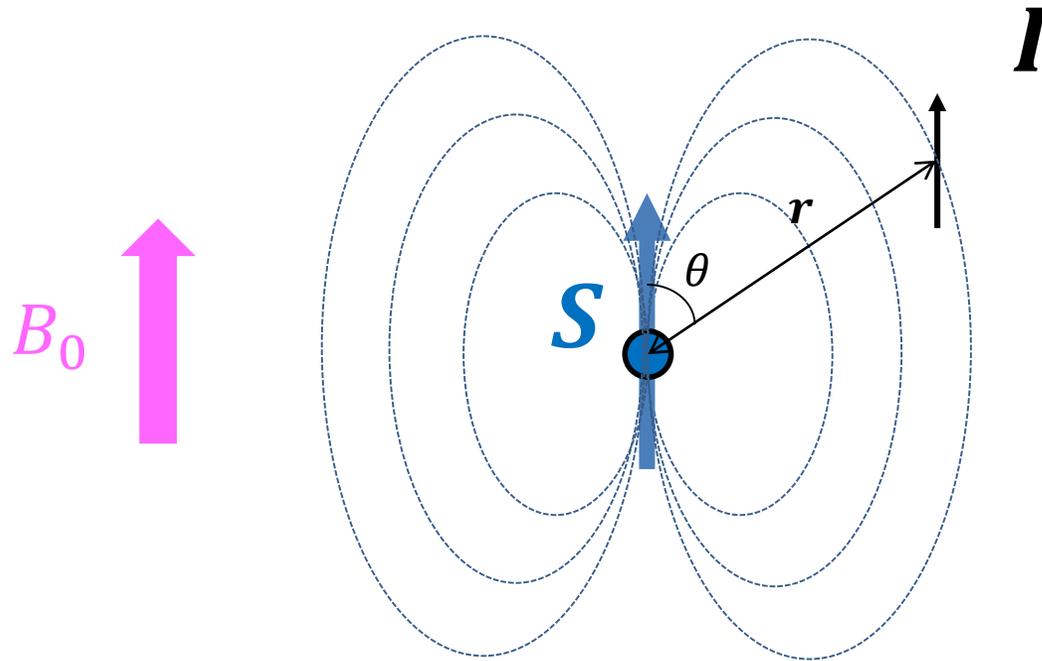
EPR spectroscopy gives access to electron spin environment using pulse sequences such as ENDOR, ELDOR, DEER, ESEEM, ...

**Can we do this with quantum-limited spectrometer ?**



Davies ENDOR with superc.  $\mu$ resonator  
Sigillito et al., Nature Nano (2017)

# Electron Spin Echo Envelope Modulation (ESEEM)



Electron spin  $S=1/2$   
Nuclear spin  $I=1/2$

Dipolar Hyperfine Constant

$$T = \frac{\mu_0}{4\pi} \frac{g_e g_n \beta_e \beta_n}{hr^3}$$

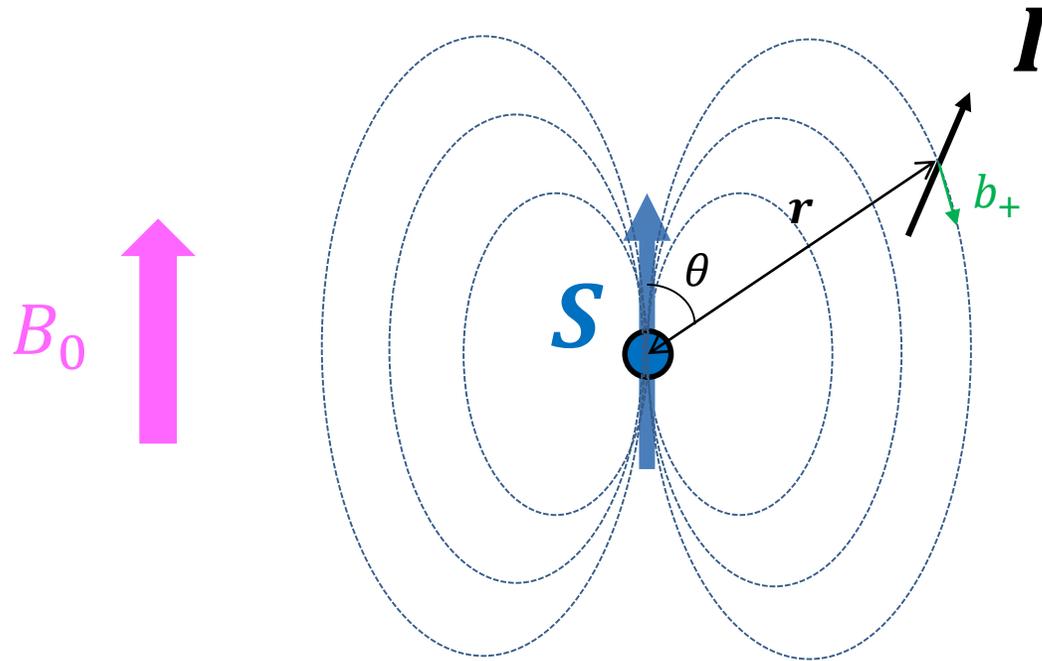
$$A = T(3 \cos^2 \theta - 1)$$

$$B = 3T \sin \theta \cos \theta$$

Dipolar hyperfine Hamiltonian

$$H = \omega_e S_z + \omega_n I_z + A S_z I_z + B S_z I_x$$

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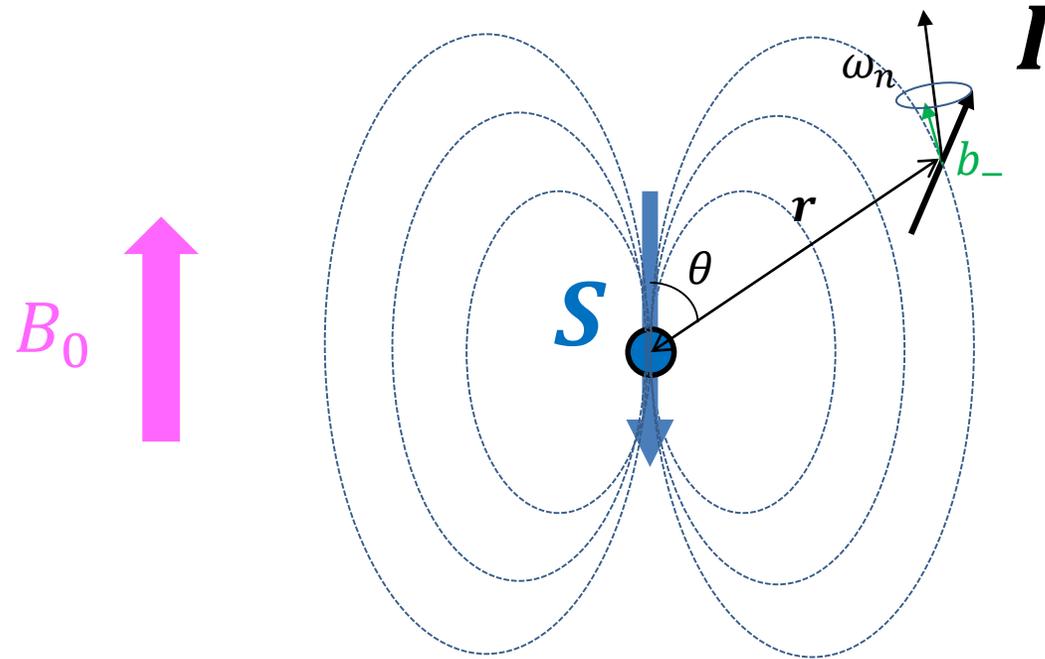
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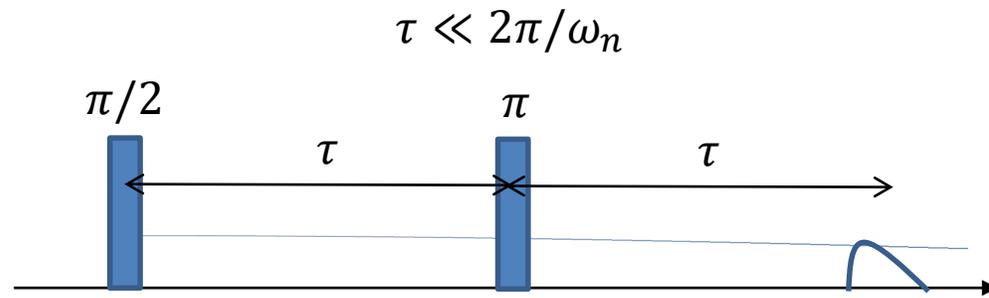
$$T = \frac{\mu_0 g_e g_n \beta_e \beta_n}{4\pi hr^3}$$

$$A = T(3 \cos^2 \theta - 1)$$

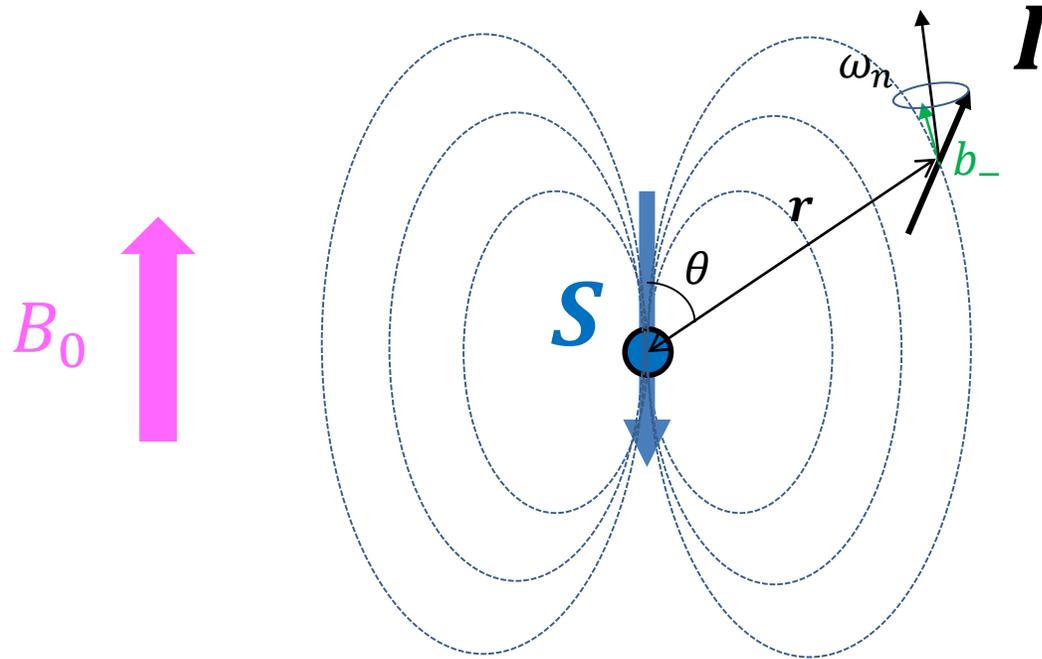
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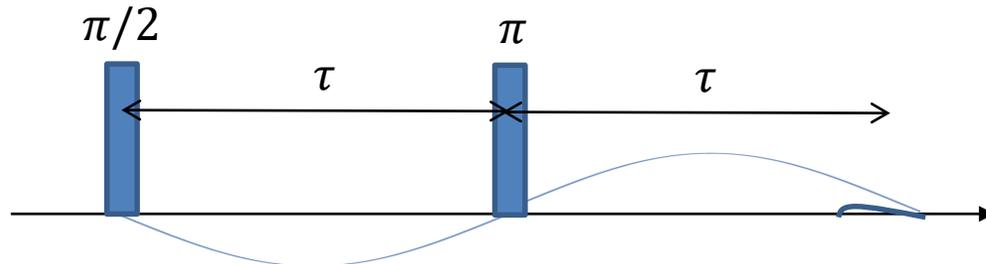
$$A = T(3 \cos^2 \theta - 1)$$

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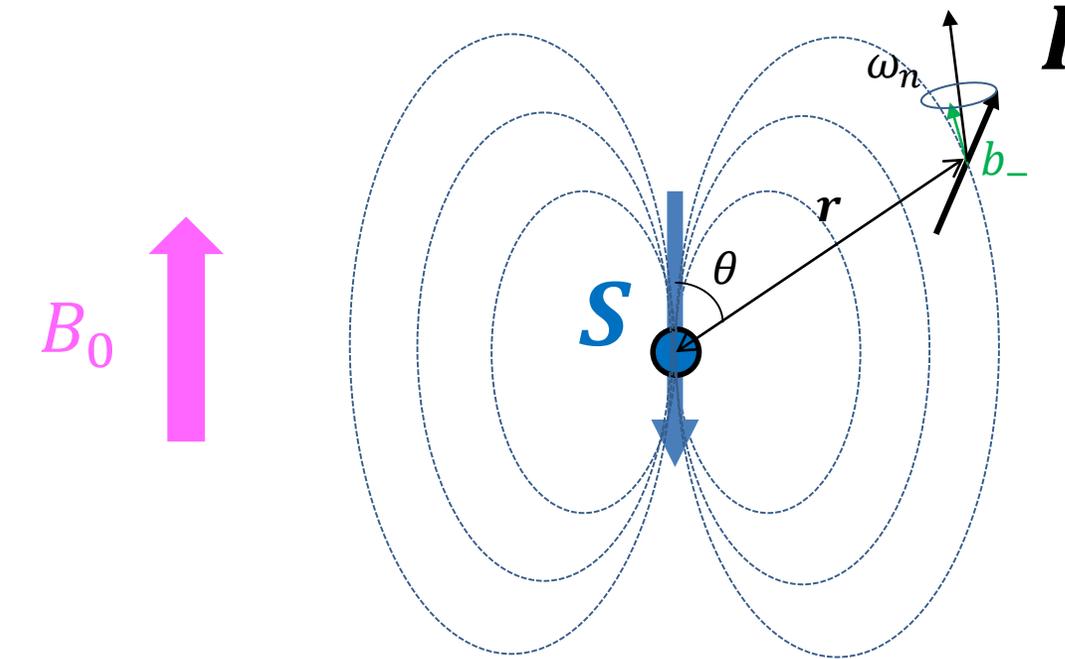
Dipolar hyperfine Hamiltonian

$$H = \omega_e S_z + \omega_n I_z + A S_z I_z + B S_z I_x$$

$\tau = \pi/\omega_n$  Suppressed echo



# Electron Spin Echo Envelope Modulation (ESEEM)



Electron spin  $S=1/2$   
Nuclear spin  $I=1/2$

Dipolar Hyperfine Constant

$$T = \frac{\mu_0}{4\pi} \frac{g_e g_n \beta_e \beta_n}{hr^3}$$

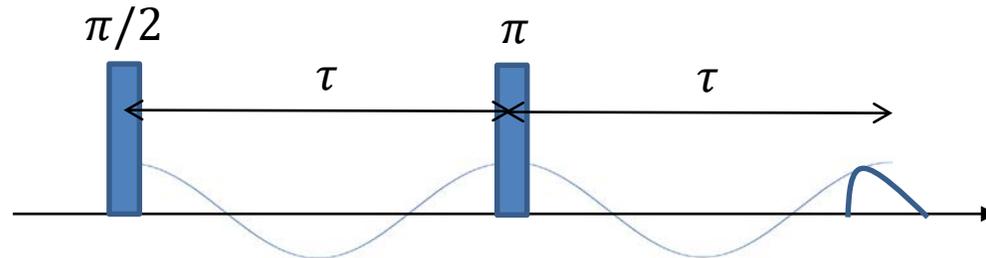
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Dipolar hyperfine Hamiltonian

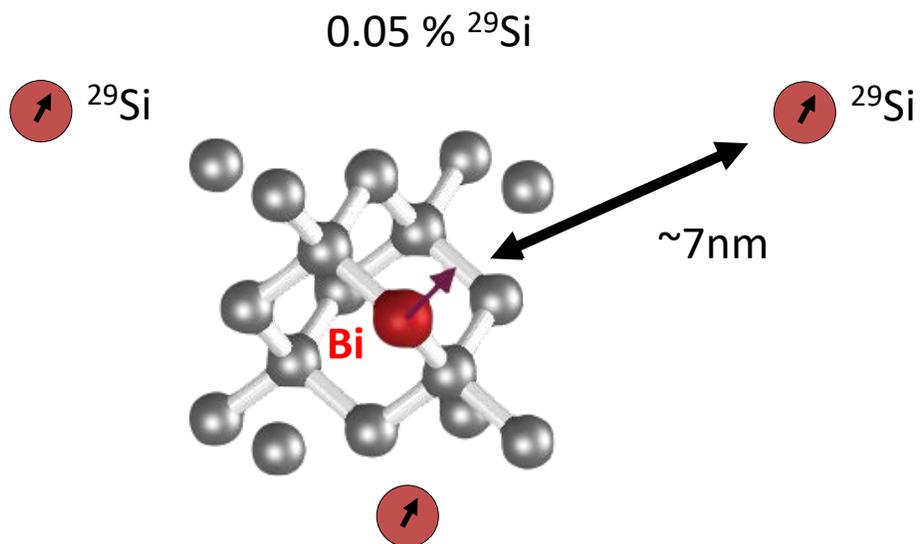
$$H = \omega_e S_z + \omega_n I_z + A S_z I_z + B S_z I_x$$

$\tau = 2\pi/\omega_n$  Echo recovery



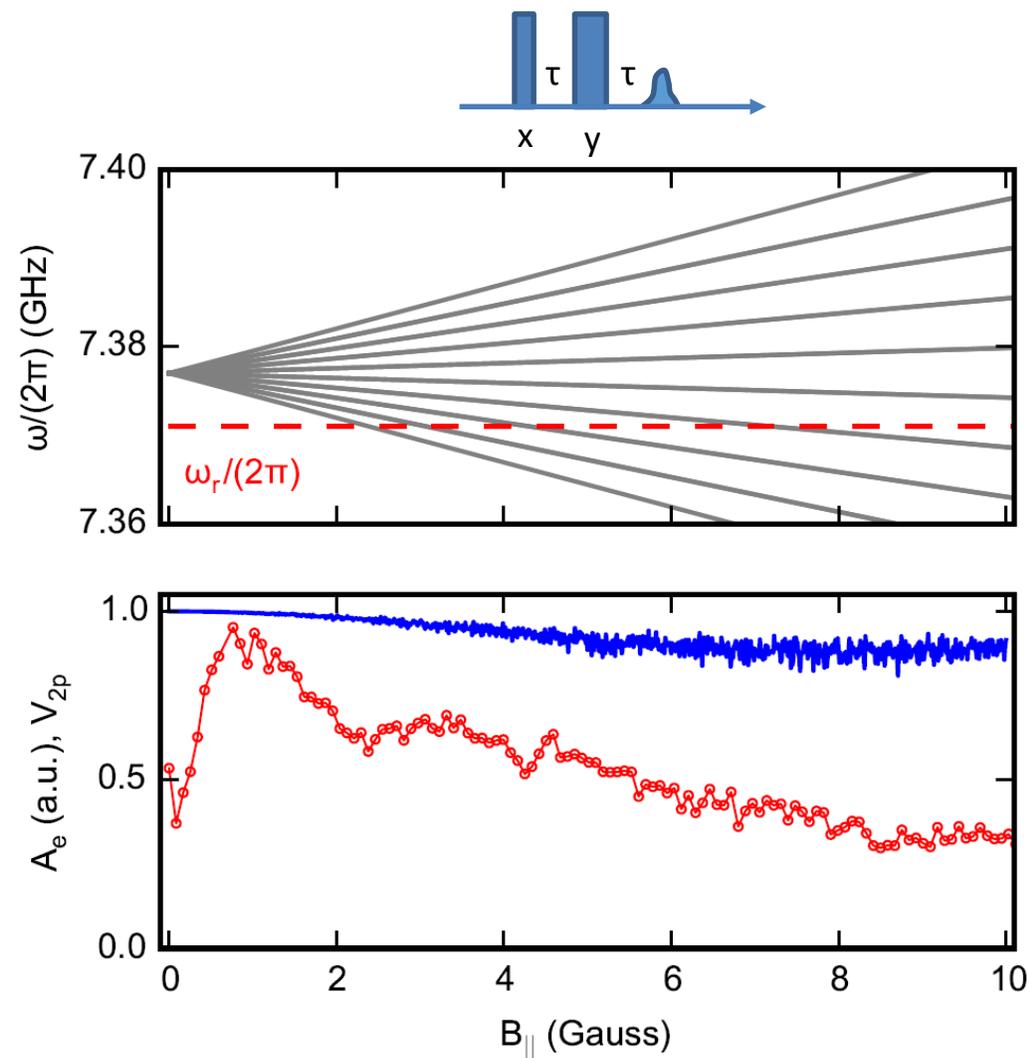
- Modulation of spin-echo envelope at  $\approx \omega_n$
- Modulation amplitude  $\sim \left(\frac{B(\theta)}{\omega_n}\right)^2$

# Detecting residual $^{29}\text{Si}$ nuclear spins through ESEEM

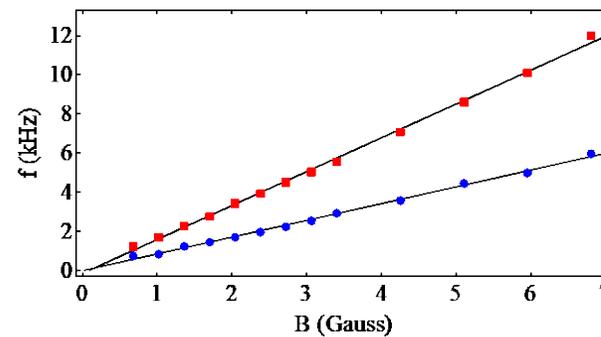
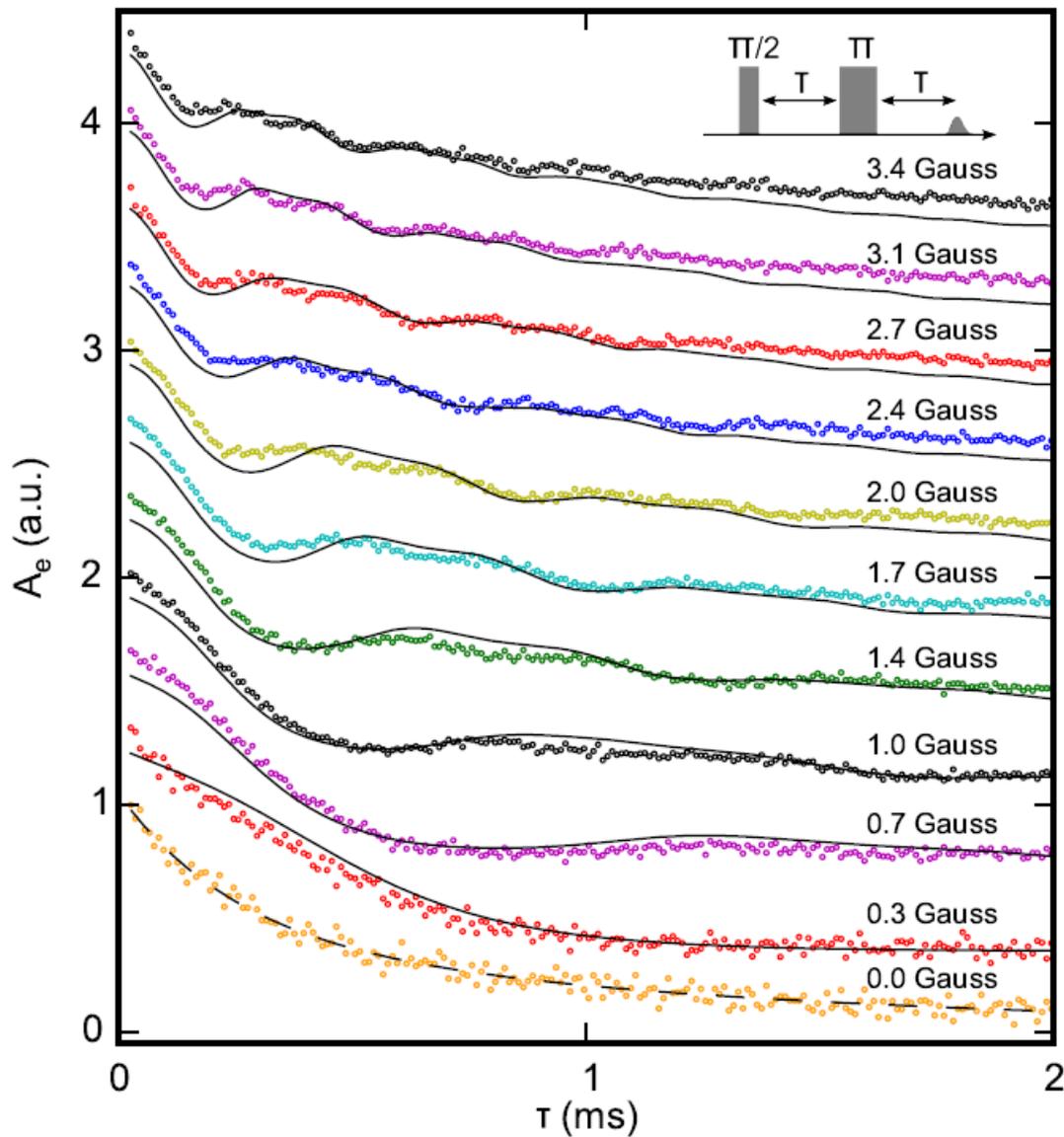


Typical  $\frac{T}{2\pi} \approx 1\text{kHz}$

Need to measure at very low  $B_0$

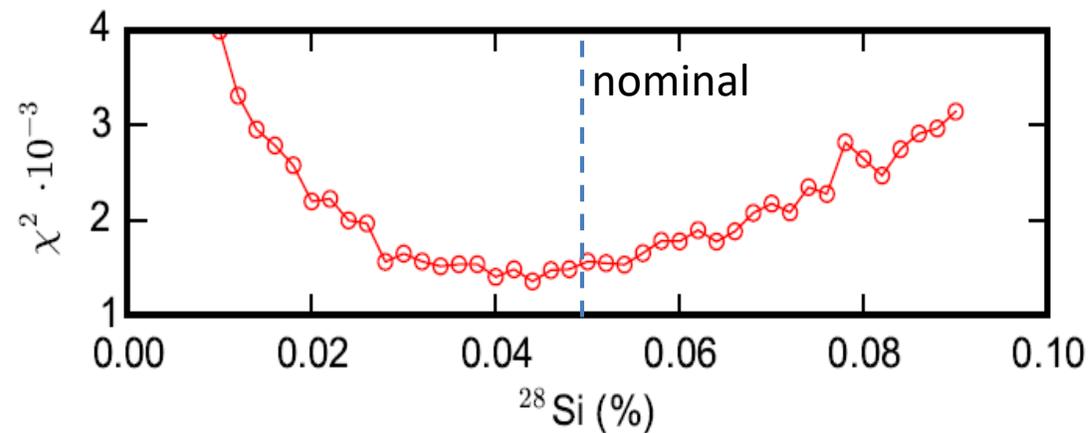


# Sensing $^{29}\text{Si}$ nuclear spins through ESEEM



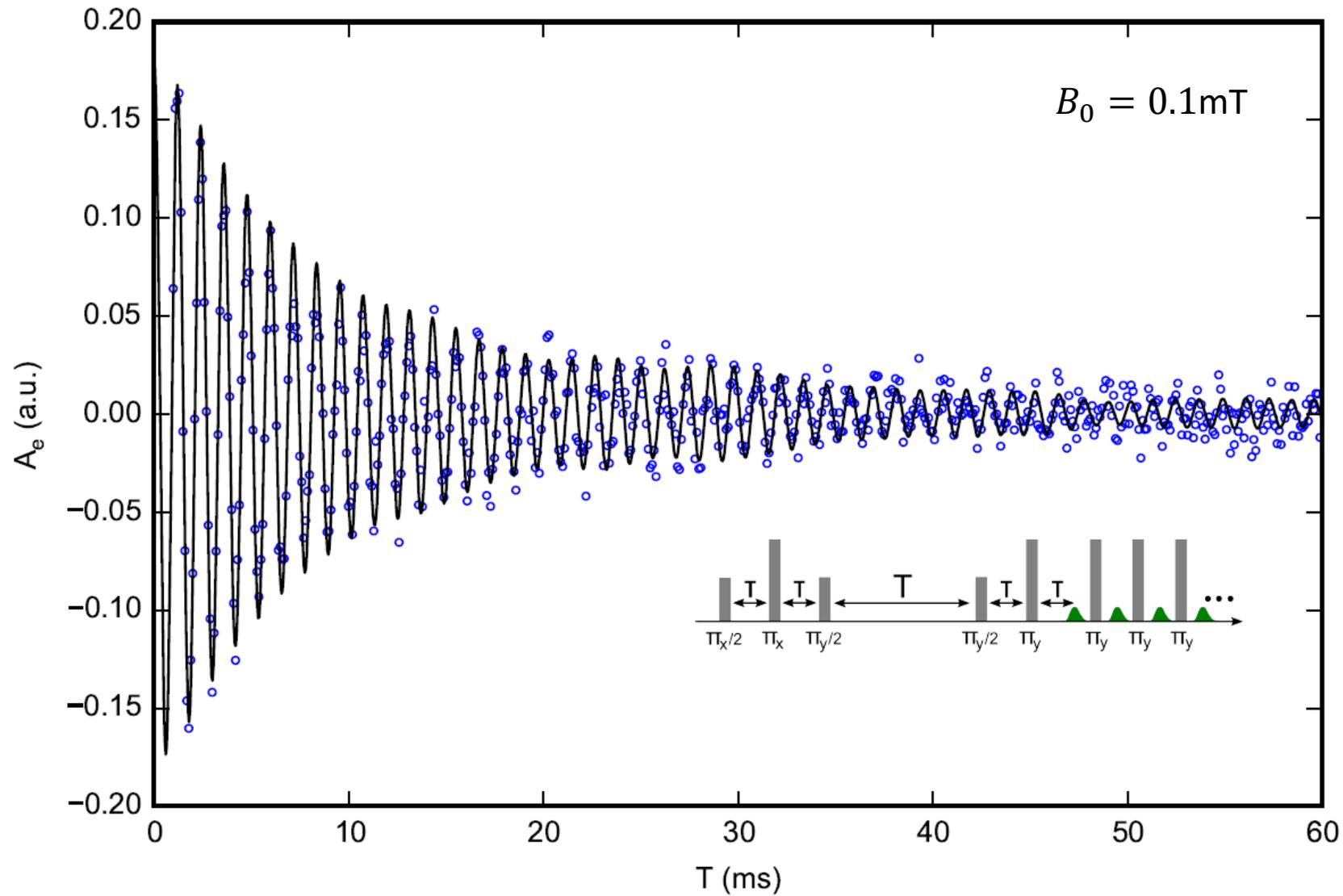
Oscillation 8.5 MHz/T  
corresponds to  $^{29}\text{Si}$   
gyromagnetic ratio

Independent confirmation of  $^{29}\text{Si}$  concentration

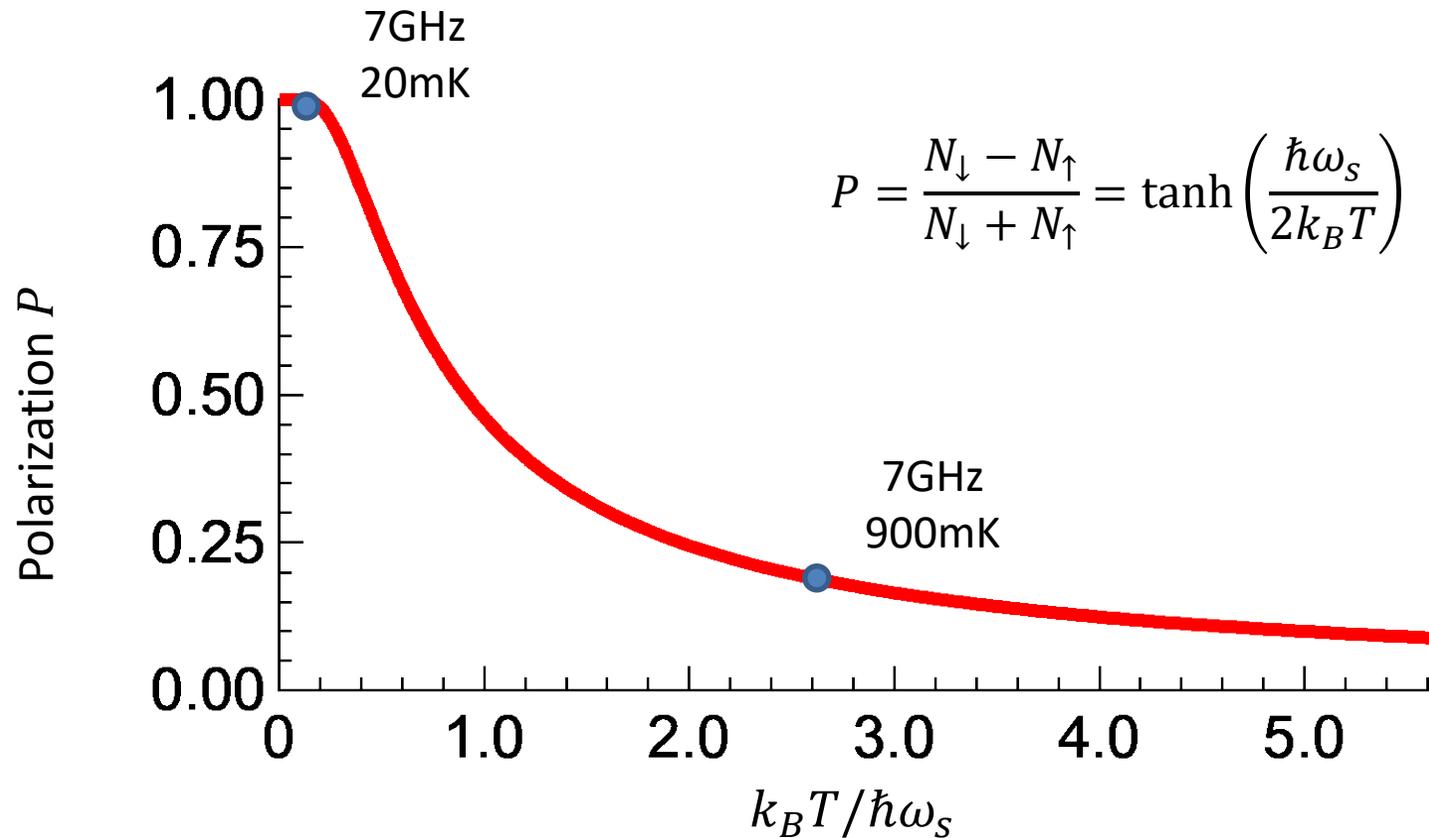


Signal comes from  $\approx 100$  Bi donor spins  
each coupled to  $\approx 10$  nuclear spins

# Beyond T2 limitation : 5-pulse ESEEM



# Circuit-QED-enhanced electron spin hyperpolarization



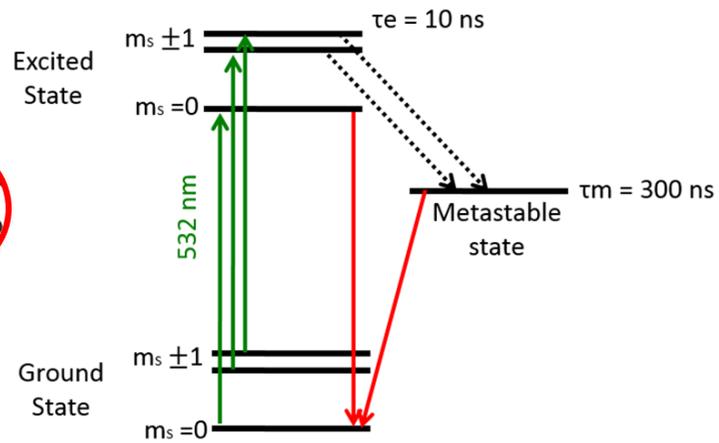
How to polarize spins beyond the Boltzmann distribution at the sample temperature  $T$  ?

# Hyperpolarization techniques

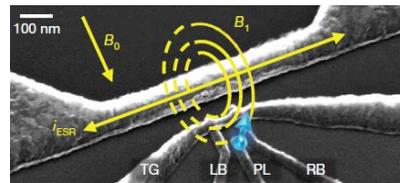
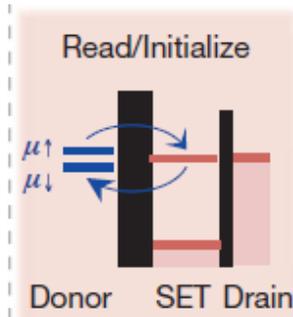
*For electron spins*

- Optical pumping

Ex: NV centers in diamond



- Spin-dependent tunneling



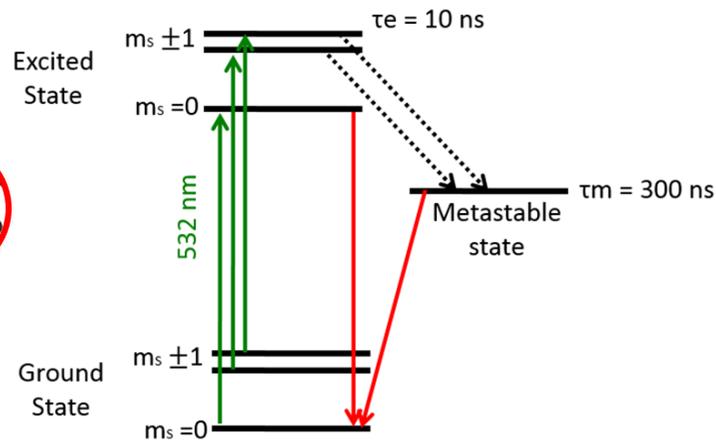
Elzerman et al. Nature (2004)  
Pla et al., Nature (2010)

# Hyperpolarization techniques

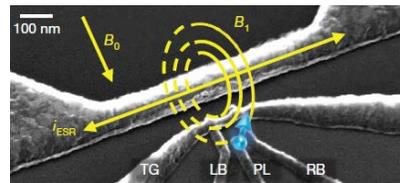
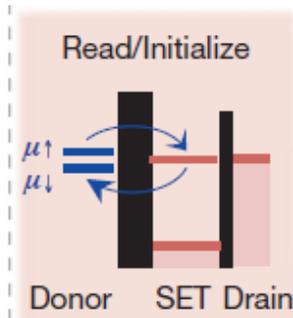
*For electron spins*

- Optical pumping

Ex: NV centers in diamond



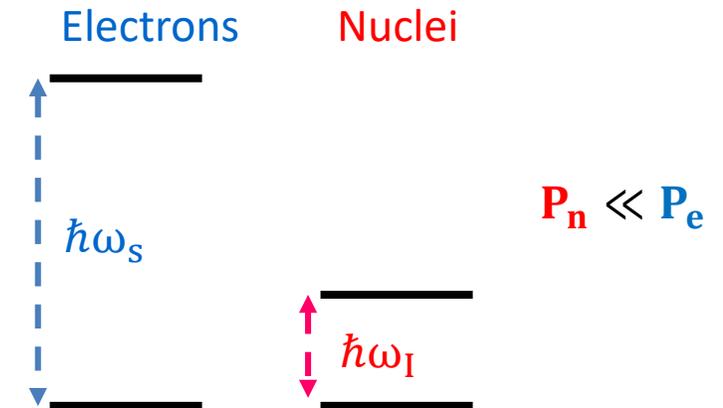
- Spin-dependent tunneling



Elzerman et al. Nature (2004)  
Pla et al., Nature (2010)

*For nuclear spins*

- Dynamical Nuclear Polarization

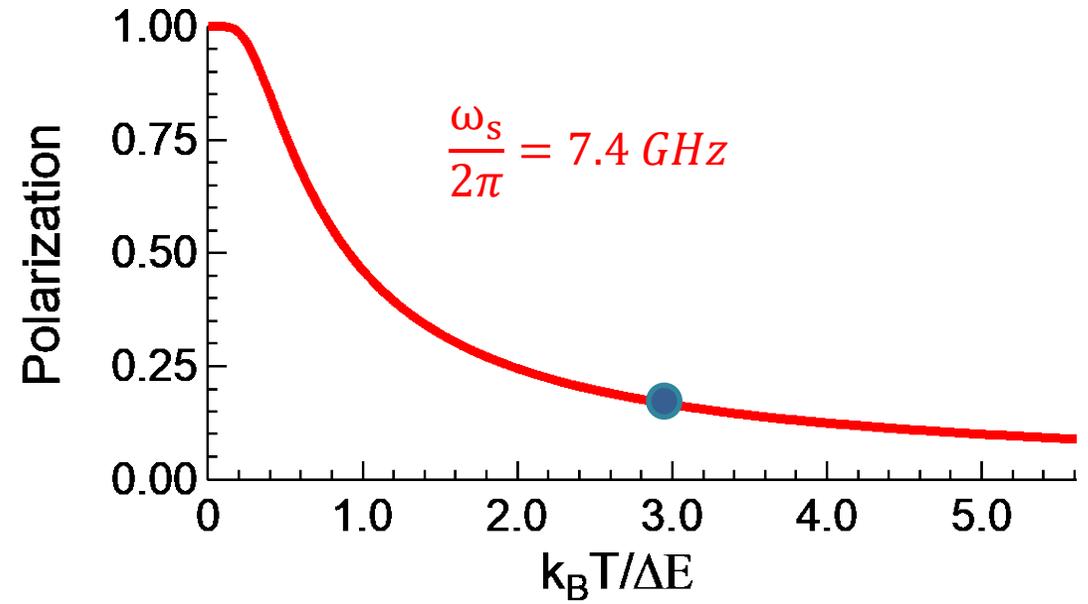
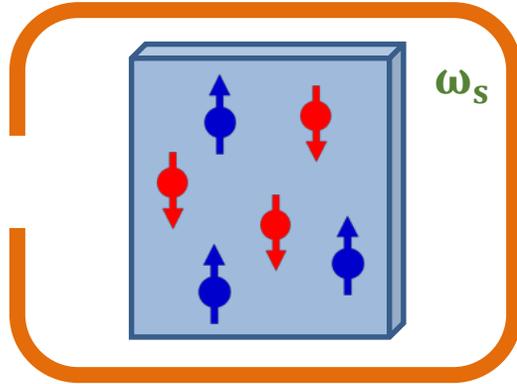


MW drive at  $\simeq \omega_s - \omega_I$    $P_n \cong P_e$

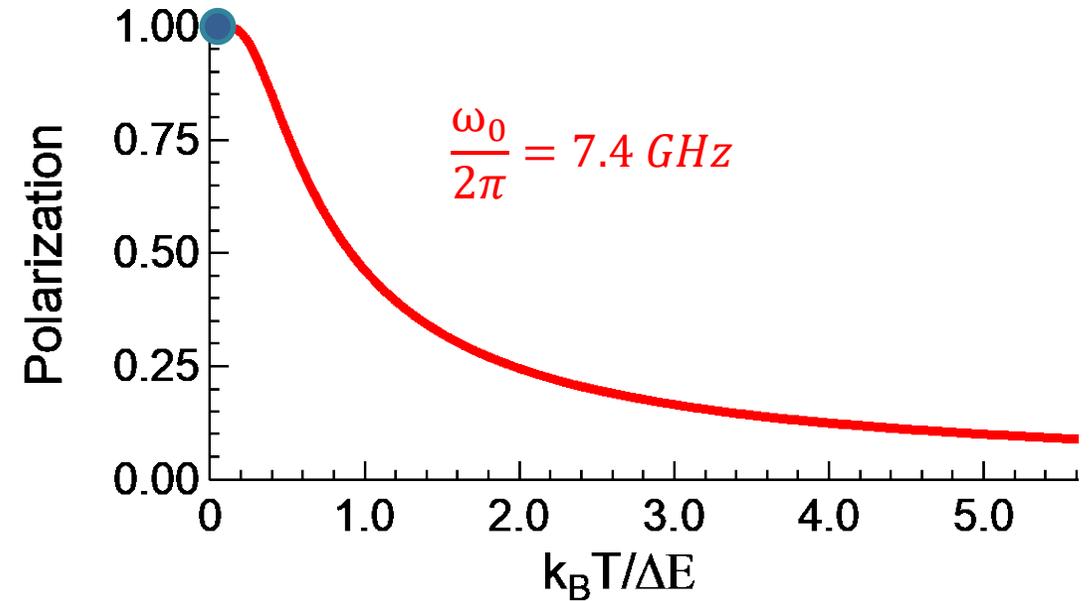
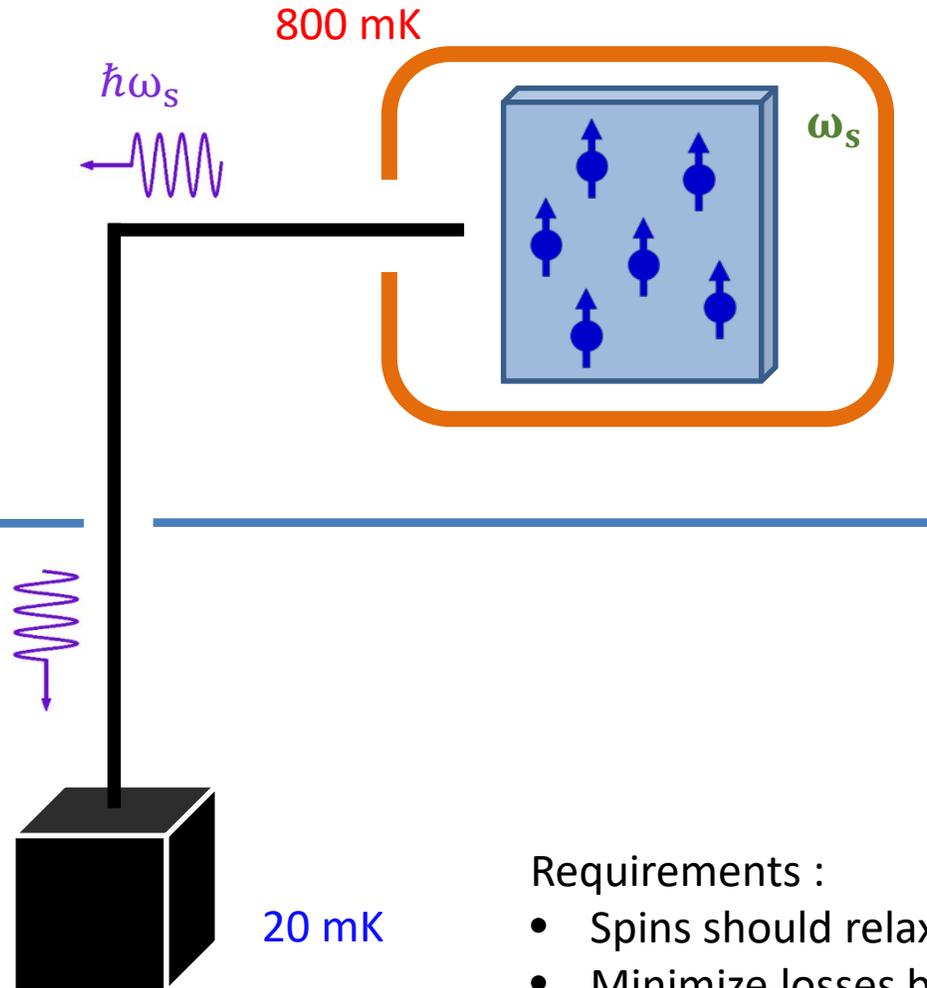
*Interest for a more general hyperpolarization scheme for electron spins*

# Hyperpolarization via radiative cooling

800 mK



# Hyperpolarization via radiative cooling



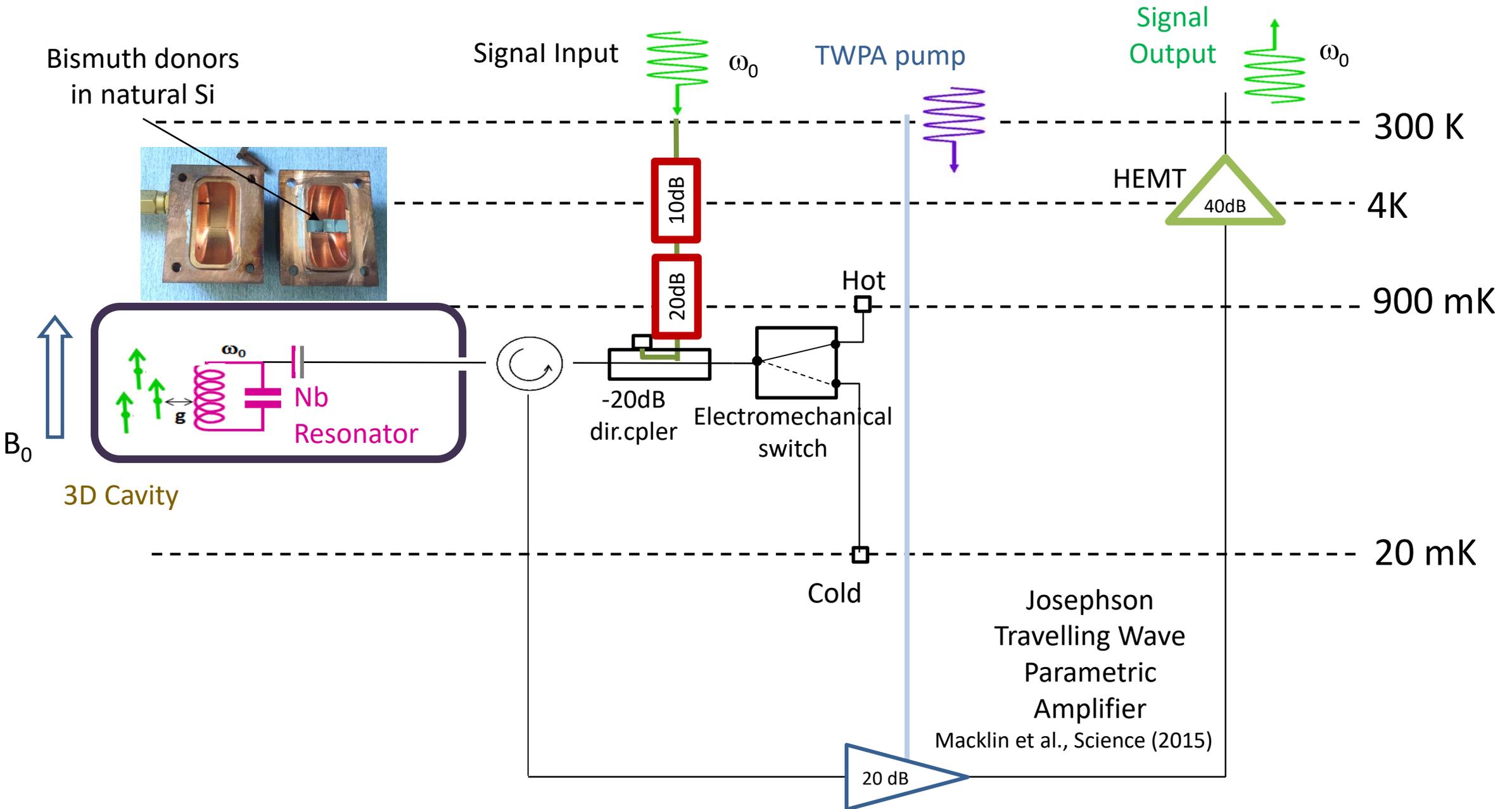
$$\frac{P_{cold}}{P_{hot}} = \frac{2\bar{n}_{800mK} + 1}{2\bar{n}_{20mK} + 1} = 4.5$$

Requirements :

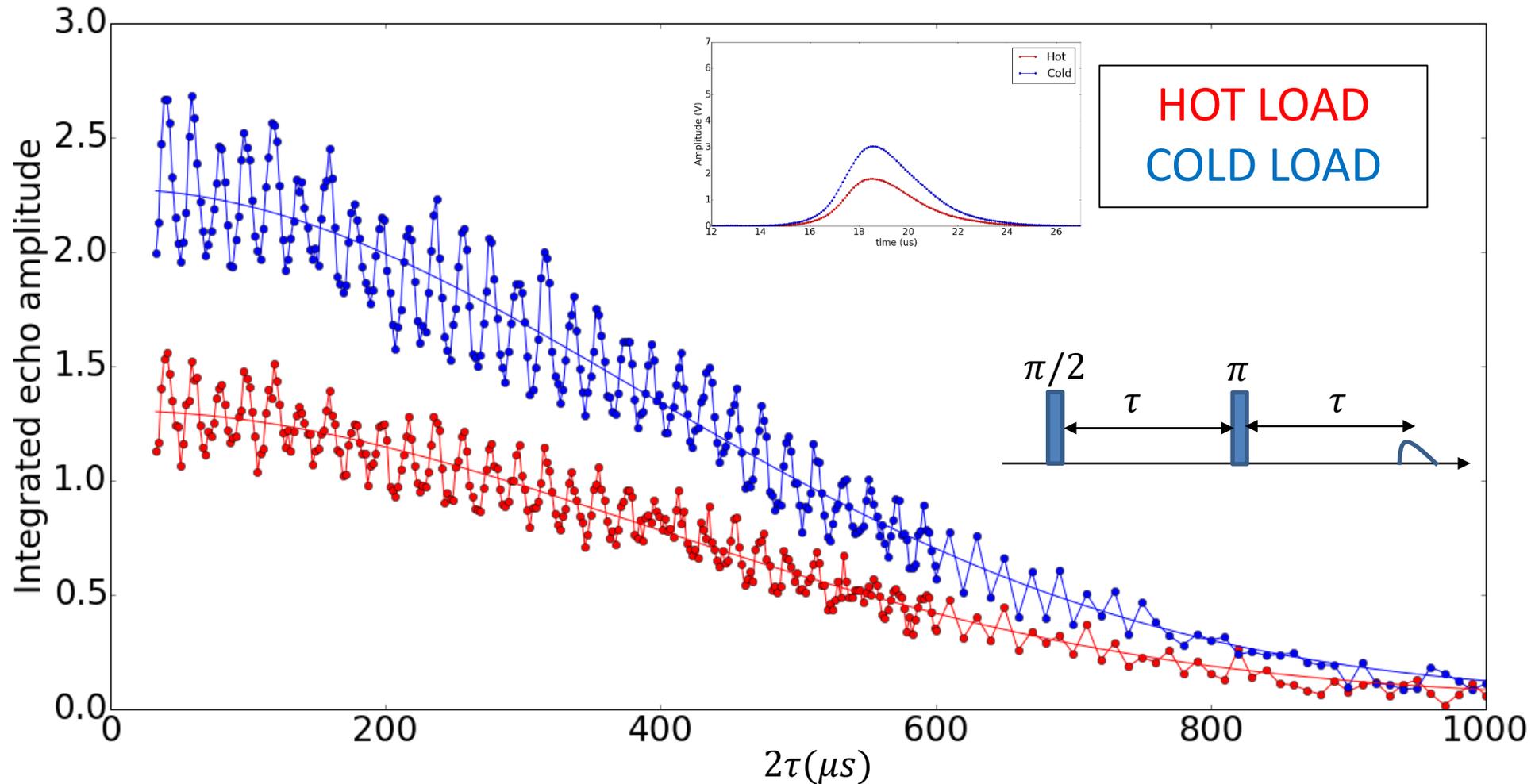
- Spins should relax radiatively (i.e. Purcell regime)
- Minimize losses between cold load and spins

Enabled by  
Circuit QED

# Hyperpolarization by radiative cooling : implementation

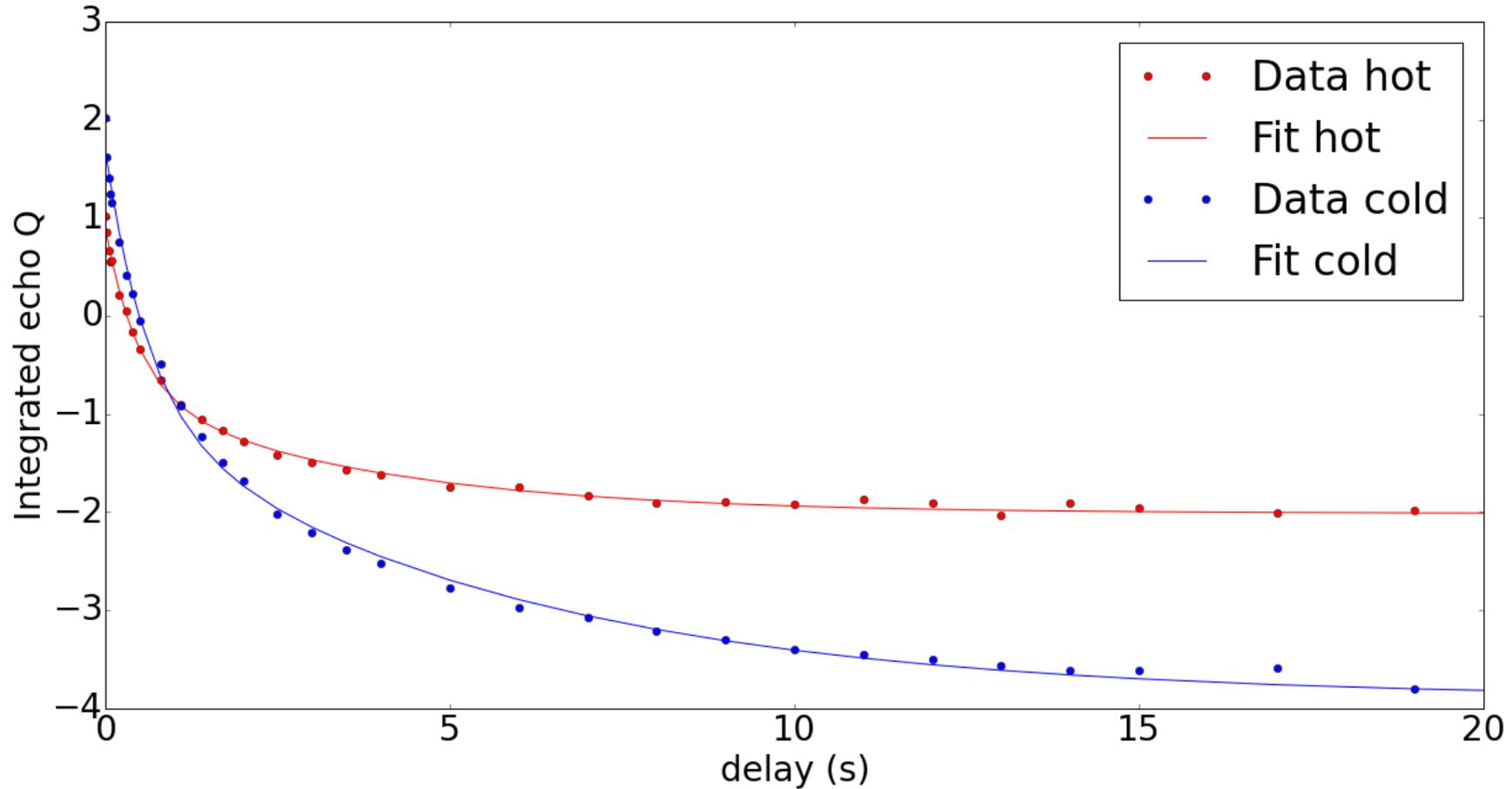


# Hyperpolarization by radiative cooling : results



- Strong ESEEM (natural abundance  $^{29}\text{Si}$ )
- Polarization increase by factor 1.9 – 2.1 when cold load connected. Less than expected due to unwanted losses

# Hyperpolarization by radiative cooling : results



- Spin relaxation time reduced by the same factor as polarization as expected

# Conclusion

- Circuit-QED-based EPR spectroscopy enables unprecedented sensitivity for inductive detection  
Demonstrated spin sensitivity of 10spin/√Hz, entering the few-spin regime  
Bienfait et al., Nature Nano (2016)  
Probst et al., APL (2017)
- Response to strain and electric field, resonator-induced decoherence, ...need to be better understood  
Mansir et al., to appear in PRL (2018)  
Pla et al., PRAppl (2018)
- Spectrometer can be used to probe electron spin environment using standard pulse EPR techniques  
Probst et al., in preparation (2018)
- The Purcell regime offers novel schemes for electron spin hyperpolarization

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P. Jamonneau  
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X. Zhou  
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