Quantum Spintronics Design
(NV centers in diamond)

Eisuke Abe

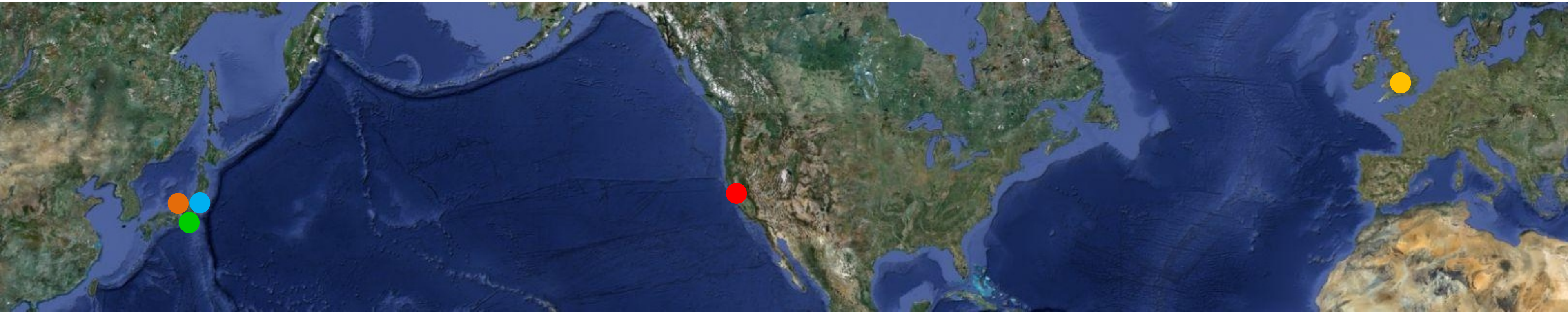
RIKEN Center for Emergent Matter Science

2019.09.04

CMD Spintronics Design Course

@Osaka University

Short CV

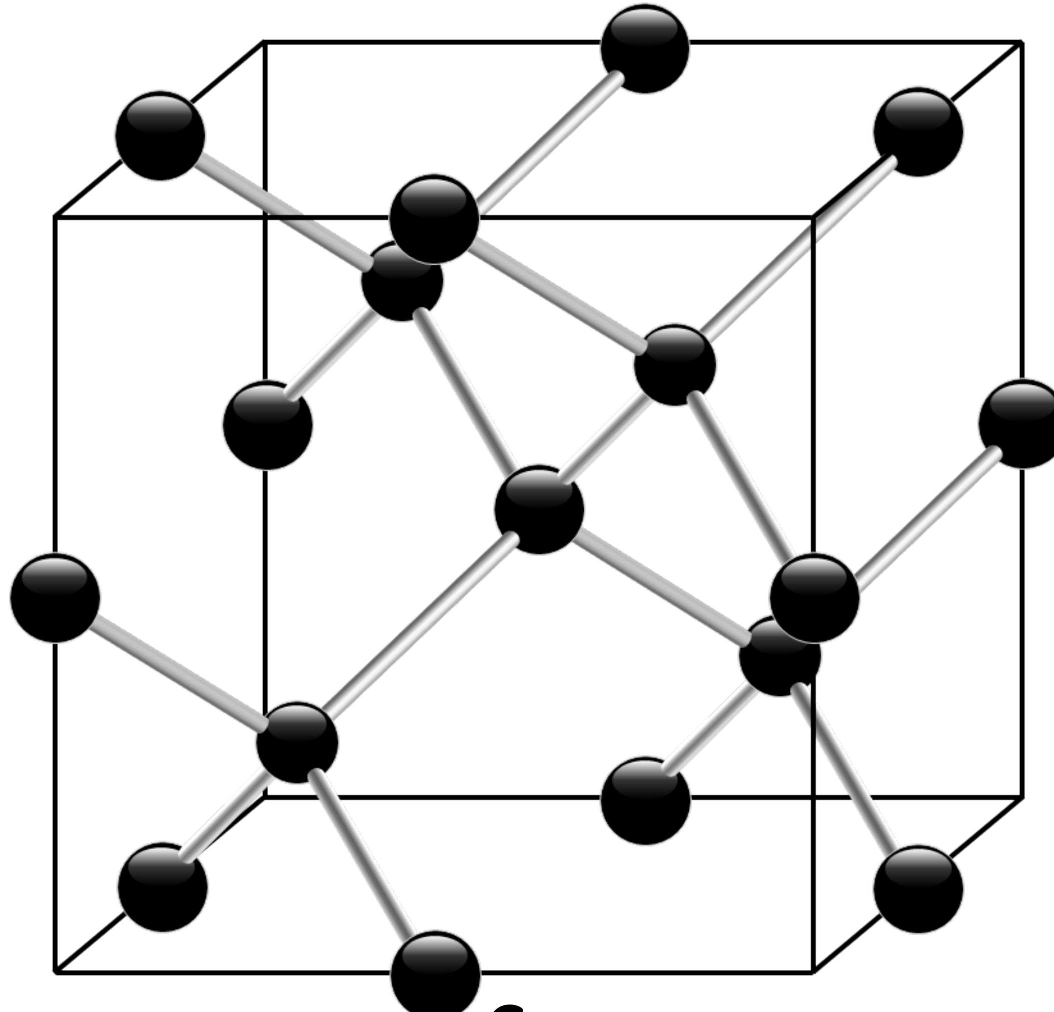


- **2001.4 – 2006.3 (Keio)** → Quantum computing (silicon)
- **2006.4 – 2009.12 (ISSP, UT)** → Quantum transport (GaAs QDs, Josephson)
- **2010.1 – 2011.6 (Oxford)** → Hybrid system (spin–cavity coupling)
- **2011.7 – 2015.3 (Stanford/RIKEN)** → Quantum network (InAs QDs)
- **2015.4 – 2019.1 (Keio)** → Quantum sensing (diamond)
- **2019.2 – Present (RIKEN)** → Quantum computing (Josephson)

Diamond envy

©Lucara Diamond

1109 carats, $70M



ρ_N = 1.77 x 10^{23} cm^{-3}

Diamond NV

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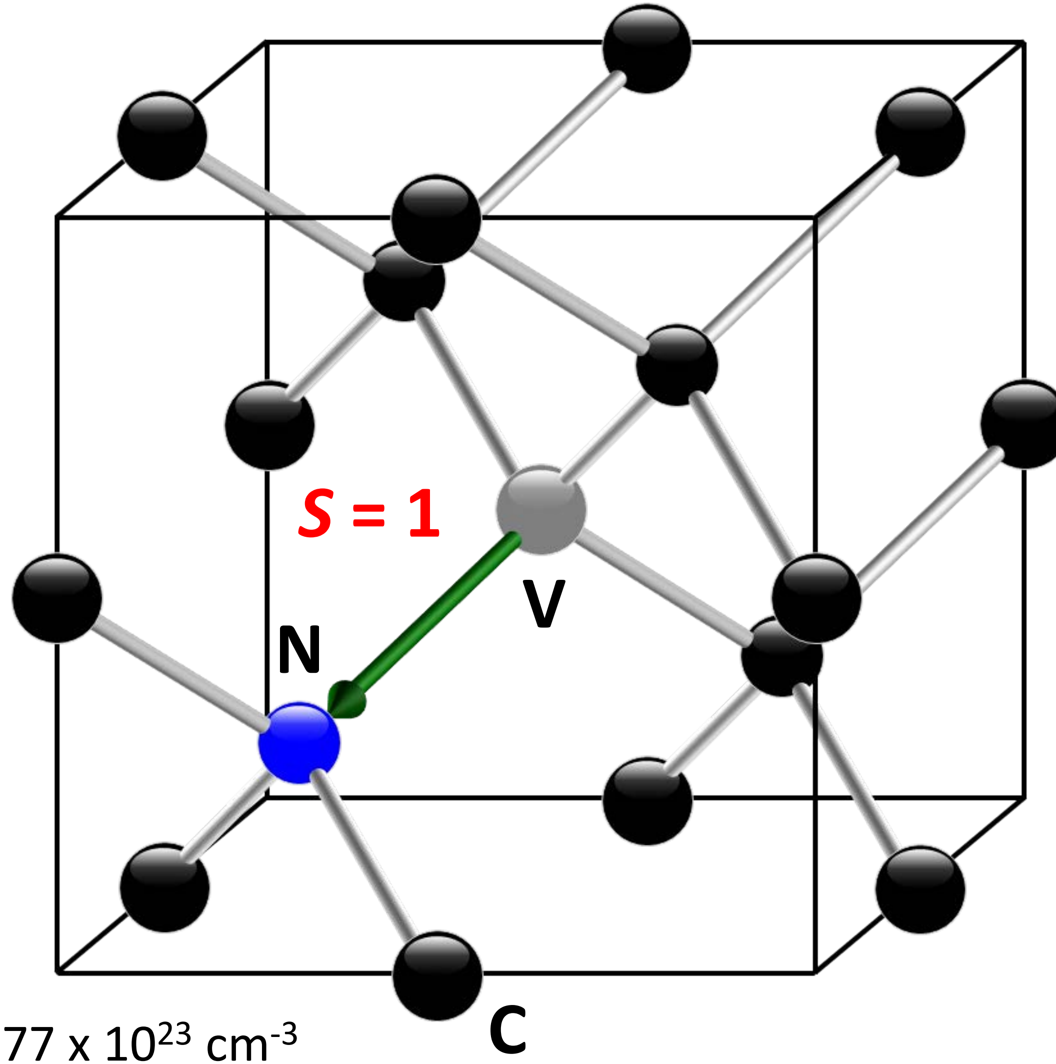
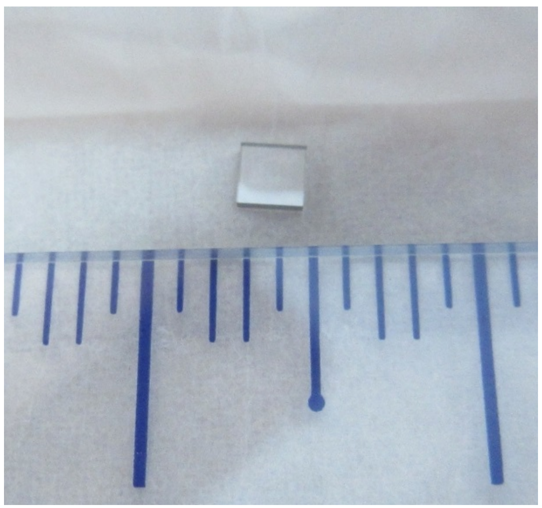
1109 carats, \$70M

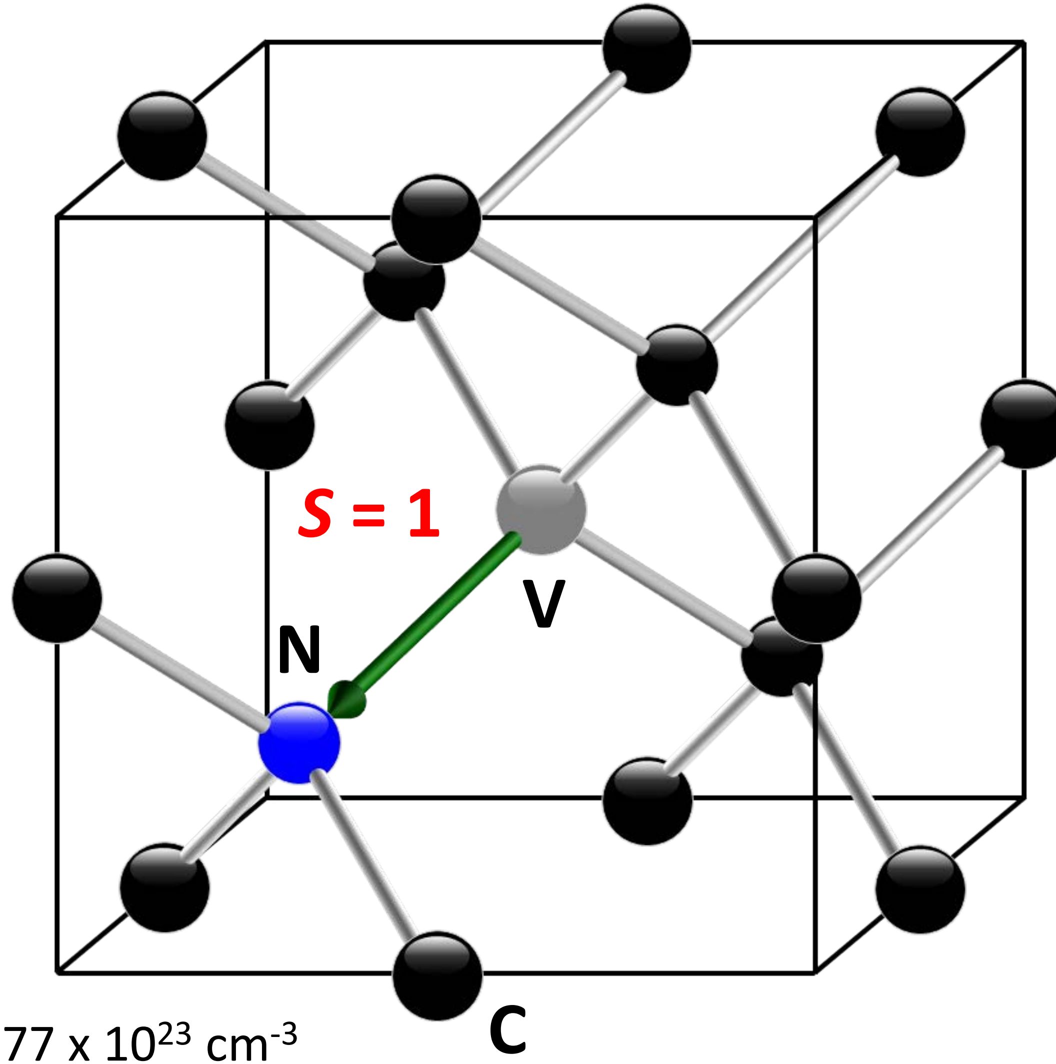


$\rho_N = 1.77 \times 10^{23}\ cm^{-3}$

Diamond NV



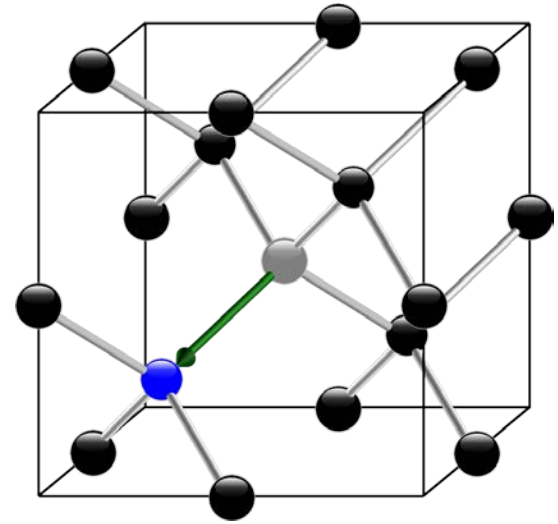
2^2 x 0.5 mm^3, $700 (E6)
[N] < 5 ppb, [NV] < 0.03 ppb



ρ_N = 1.77 x 10^{23} cm^{-3}

Why a single NV spin?

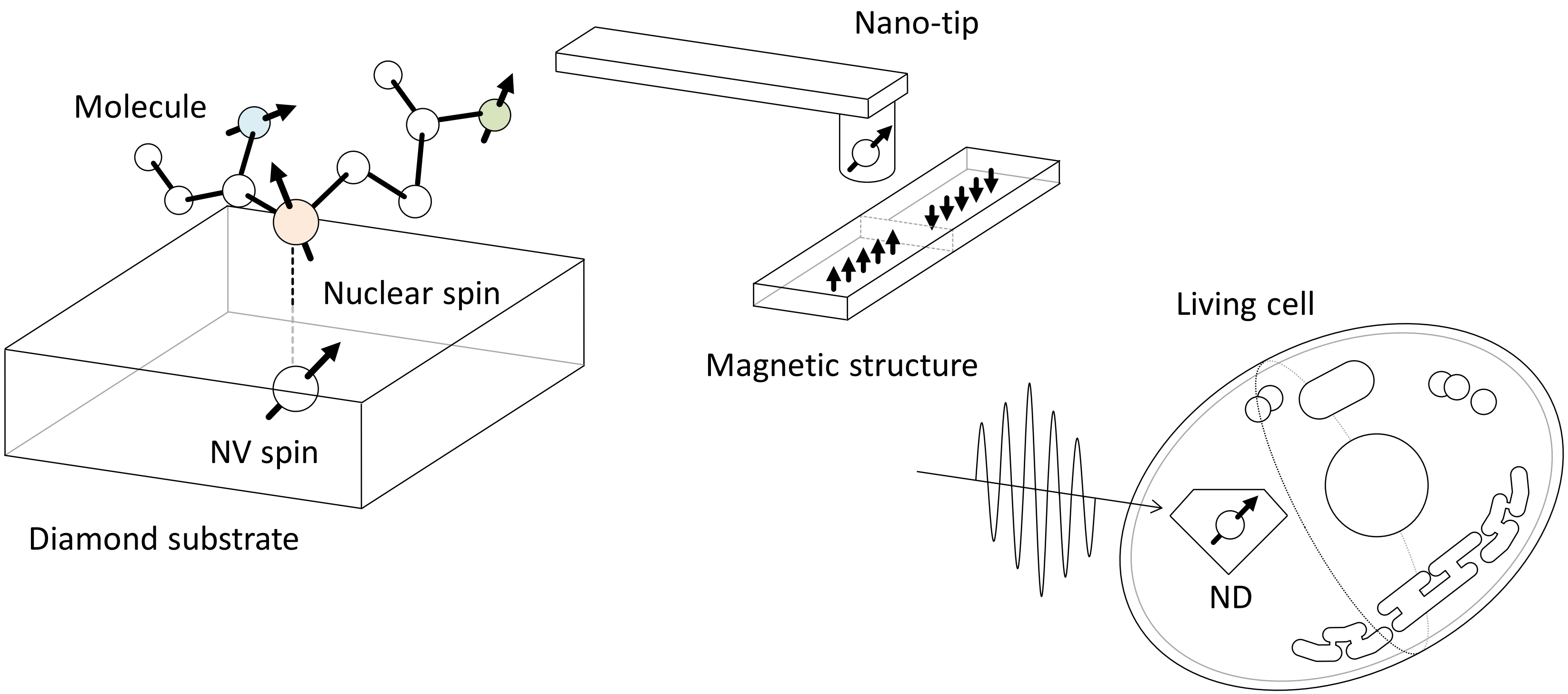
- Optically active (init. & readout)
- Microwave spin control
- High coherence (LT–RT–500 K)



⇨ **Quantum sensing & Quantum network**

[**Biology**] Annu. Rev. Phys. Chem. **65**, 83 (2014) Schirhagl *et al.*
[**Magnetometry**] Rep. Prog. Phys. **77**, 056503 (2014) Rondin *et al.*
[**Magnetic resonance**] J. Mag. Res. **269**, 225 (2016) Wrachtrup *et al.*
[**Quantum technologies**] Nature Photon. **12**, 516 (2018) Awschalom *et al.*
[**Quantum internet**] Science **362**, eaam9288 (2018) Wehner *et al.*

Quantum sensing

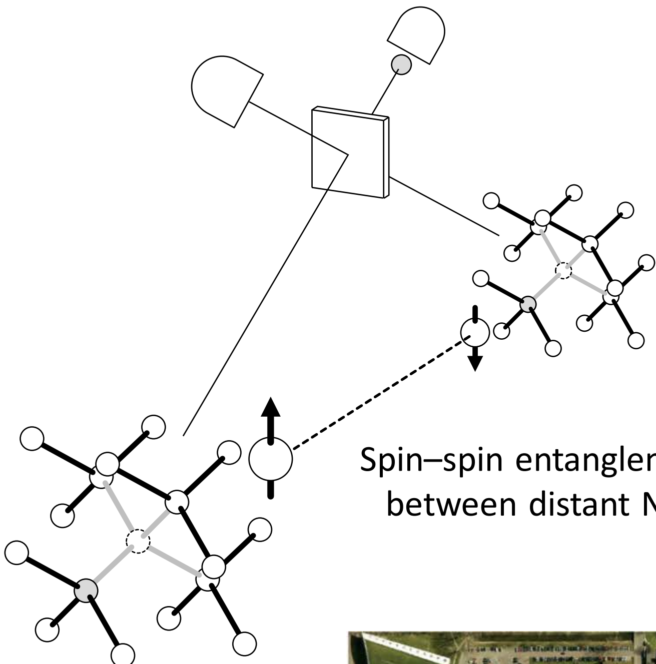


- Room T. operation
- High spatial resolution
- Nondestructive
- Various modalities

⇨

- Nano MRI
- Probe for CM systems
- Biology

Quantum network



Spin–spin entanglement between distant NVs

LETTER

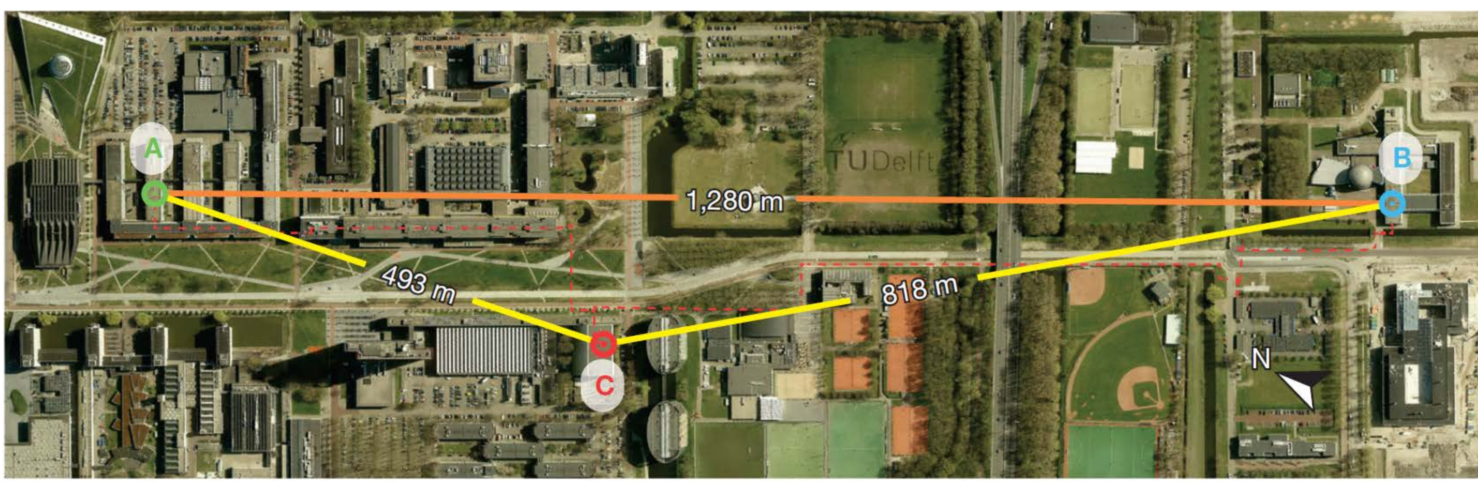
doi:10.1038/nature15759

Loophole-free Bell inequality violation using electron spins separated by 1.3 kilometres

B. Hensen[1,2], H. Bernien[1,2]†, A. E. Dréau[1,2], A. Reiserer[1,2], N. Kalb[1,2], M. S. Blok[1,2], J. Ruitenberg[1,2], R. F. L. Vermeulen[1,2], R. N. Schouten[1,2], C. Abellán[3], W. Amaya[3], V. Pruneri[3,4], M. W. Mitchell[3,4], M. Markham[5], D. J. Twitchen[5], D. Elkouss[1], S. Wehner[1], T. H. Taminiau[1,2] & R. Hanson[1,2]

Nature **526**, 682 (2015) Hensen *et al.*

Times Cited: 1210 (Google Scholar)



Outline

- **Basics of NV centers in diamond**
  - Structure
  - Optical properties
  - Spin properties

- **Quantum sensing**
  - Basics
  - Correlation spectroscopy and detection of nuclear spins
  - Ultrahigh resolution sensing
  - Determination of the position of a single nuclear spin

Outline

- **Basics of NV centers in diamond**
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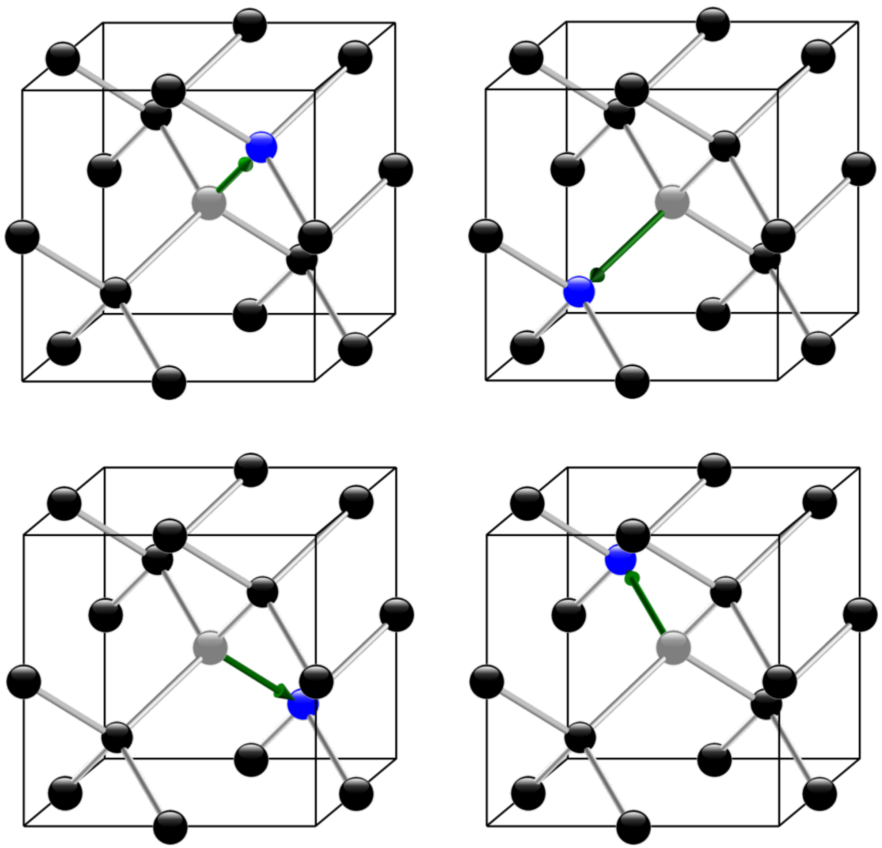
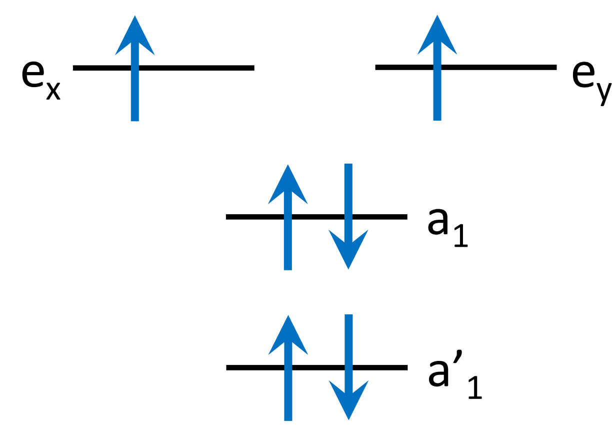
Crystal & energy level structures

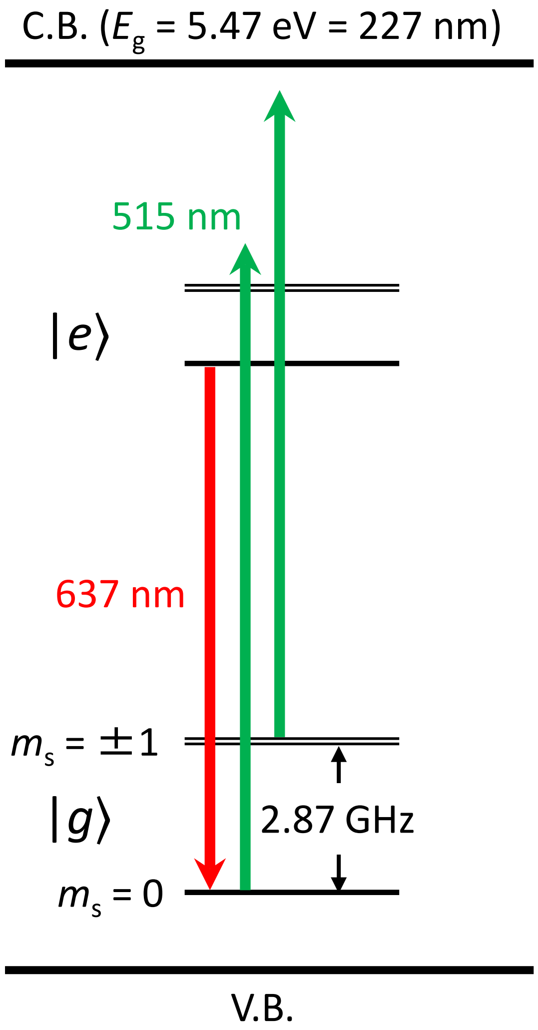
- Negatively-charged (NV^-)
- 4 sp^3 orbitals, 6 e^- (5 from the defect, 1 captured)
- C_{3v} (symmetry axis = quantization axis)

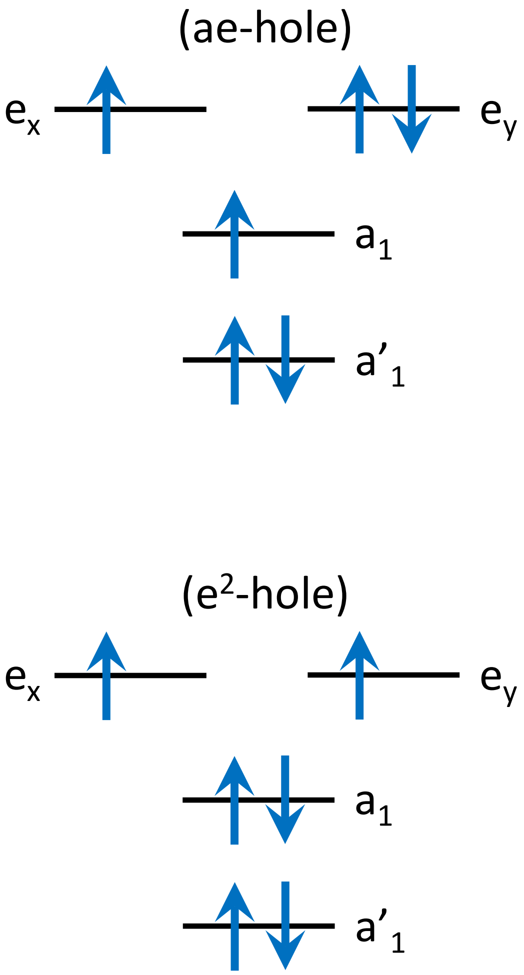


Effective spin-1 system
(e^2-hole spin-triplet)

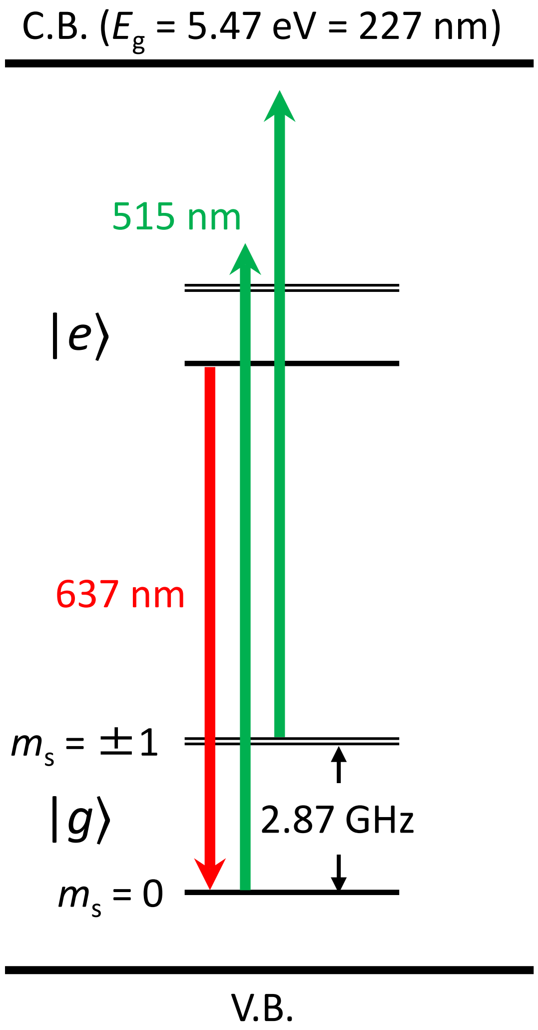


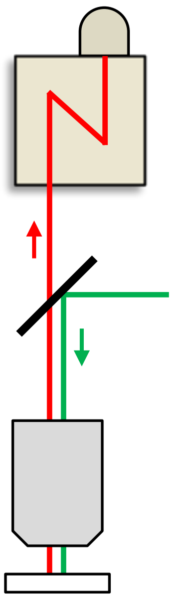
Optical transitions

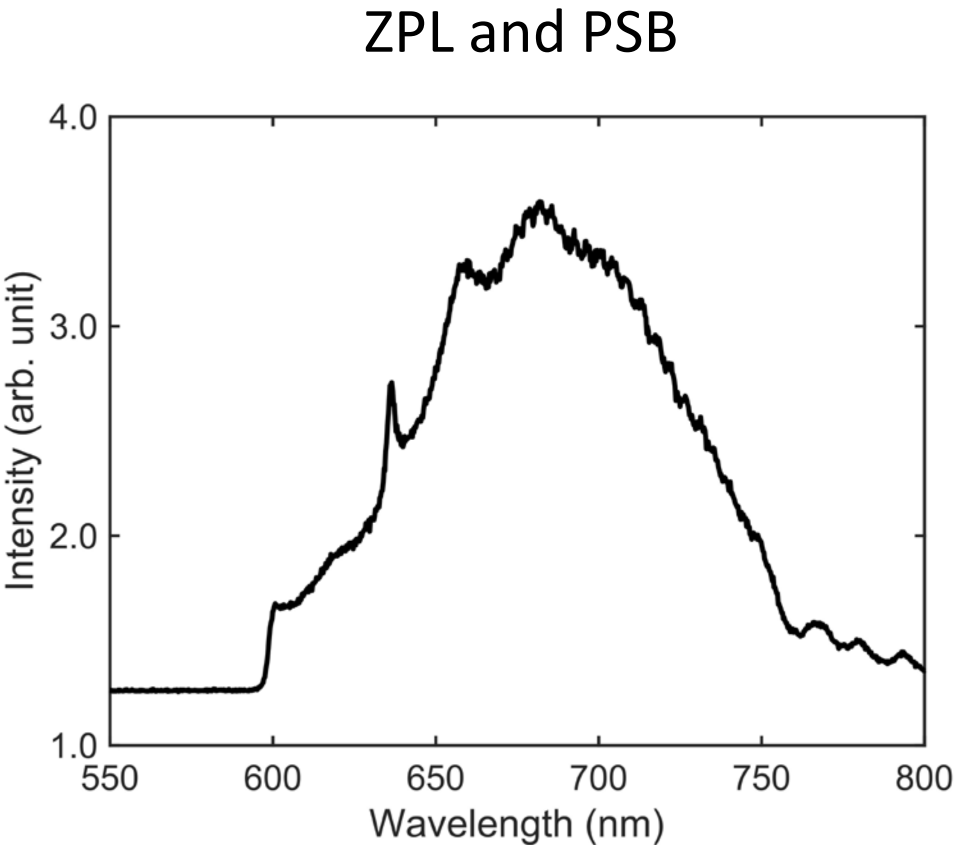




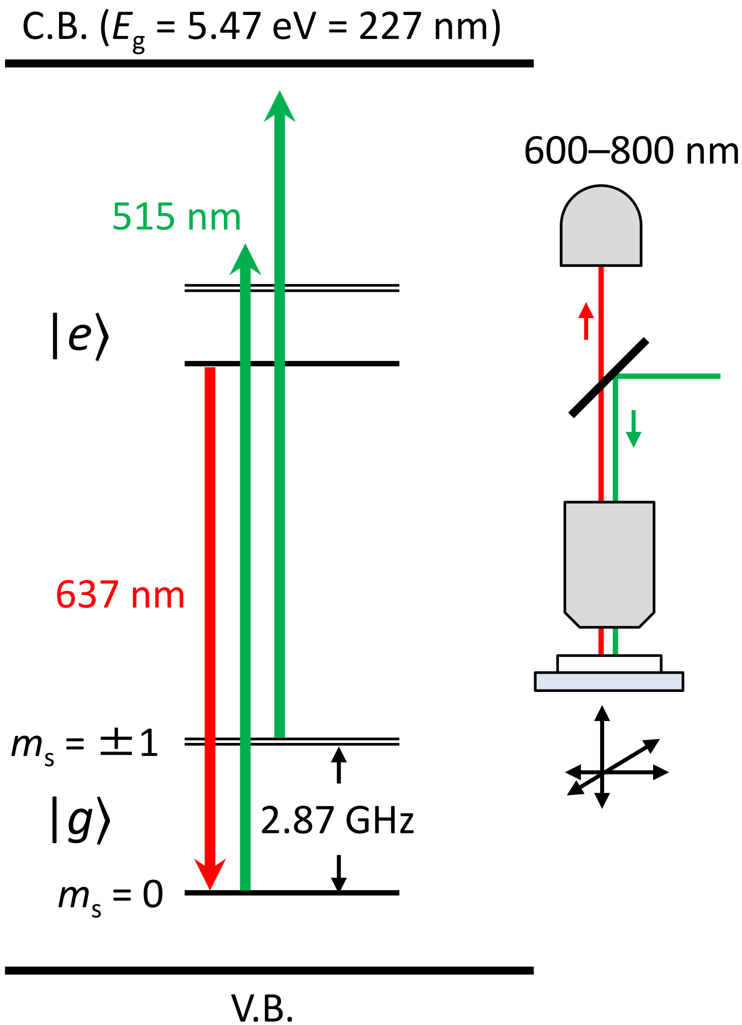
PL spectroscopy



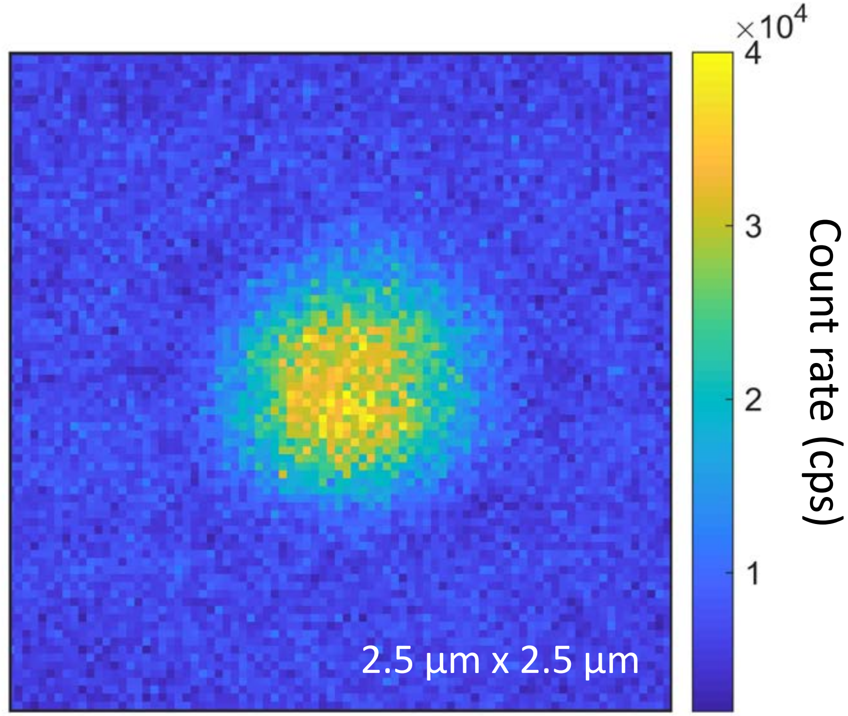




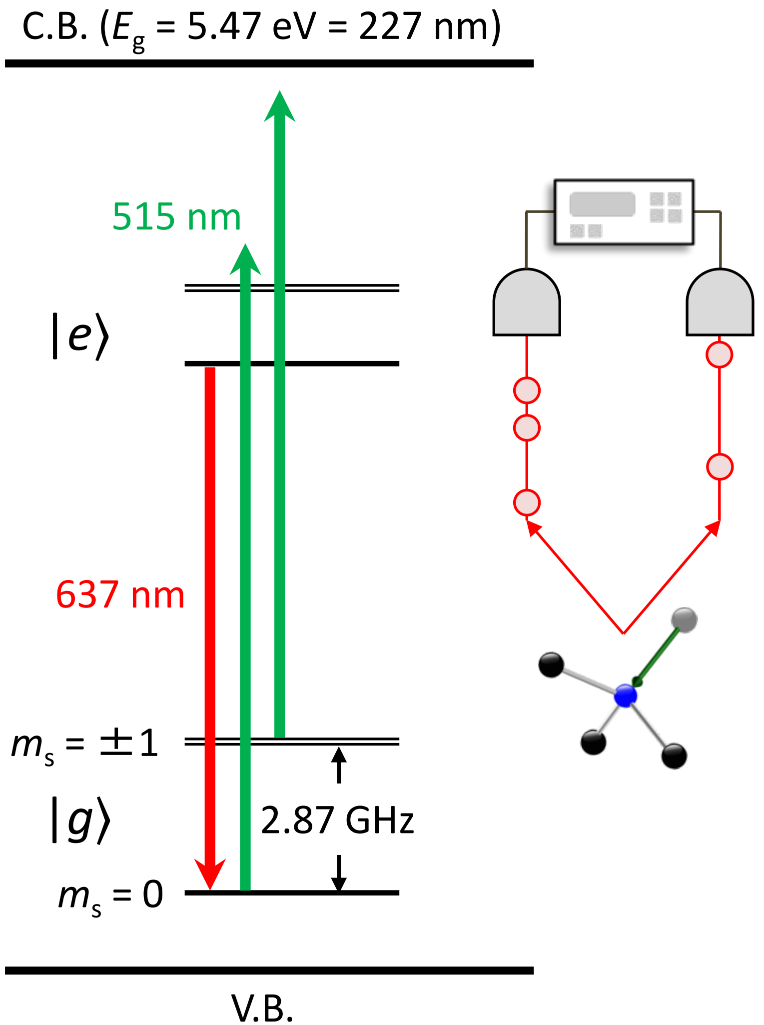
PL imaging



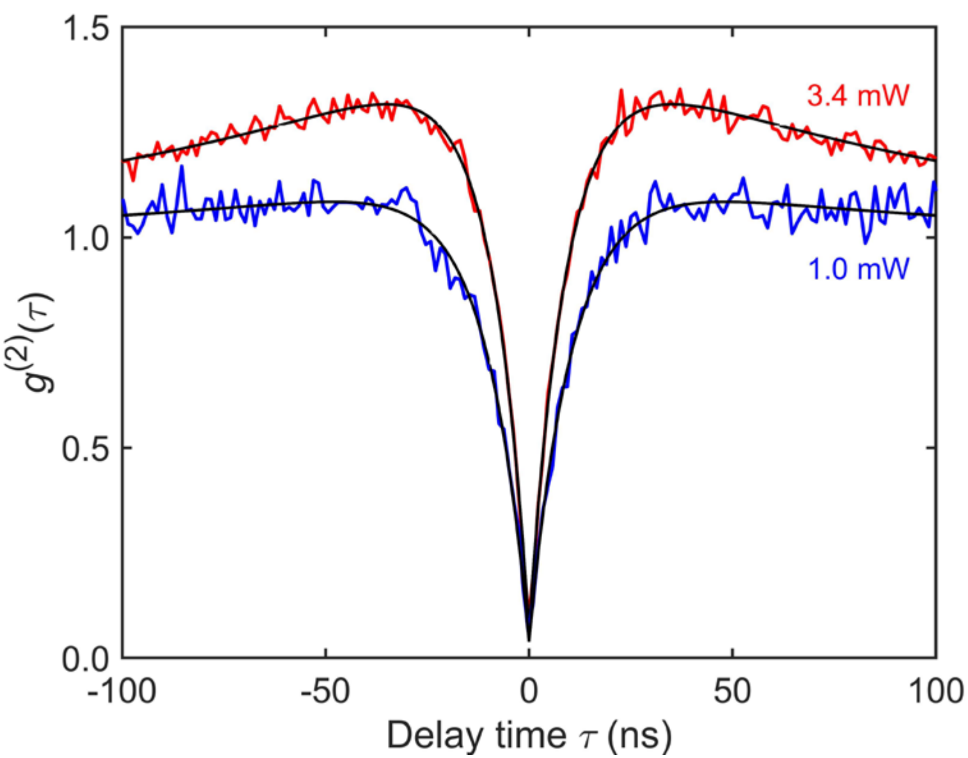
Bright spot... single NV?



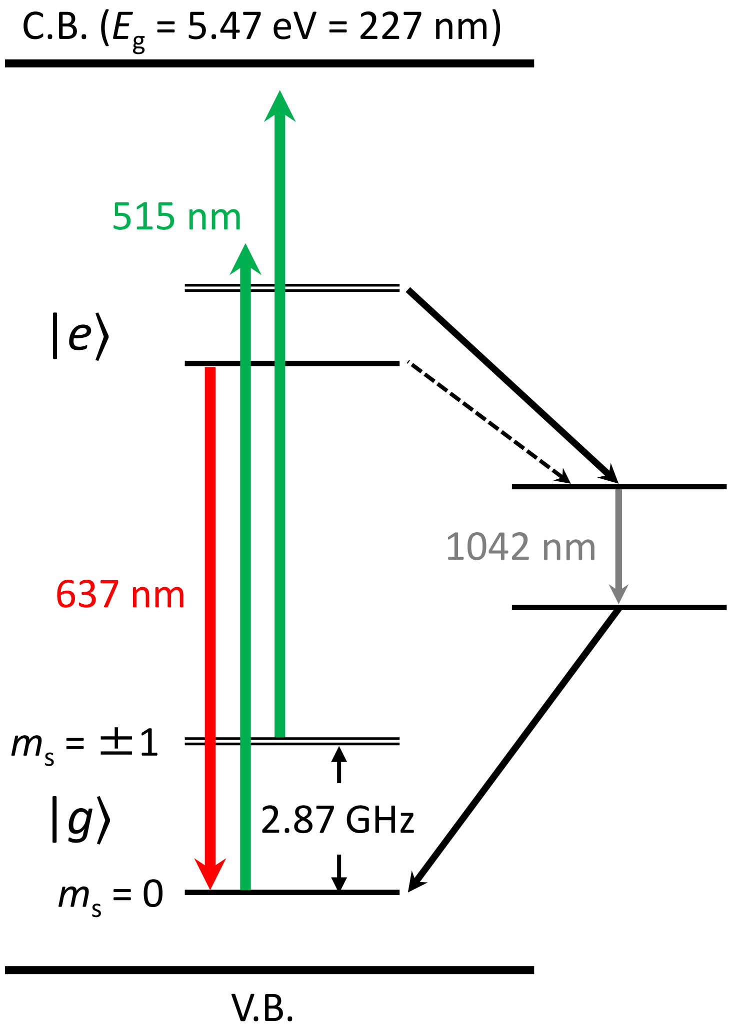
Photon statistics



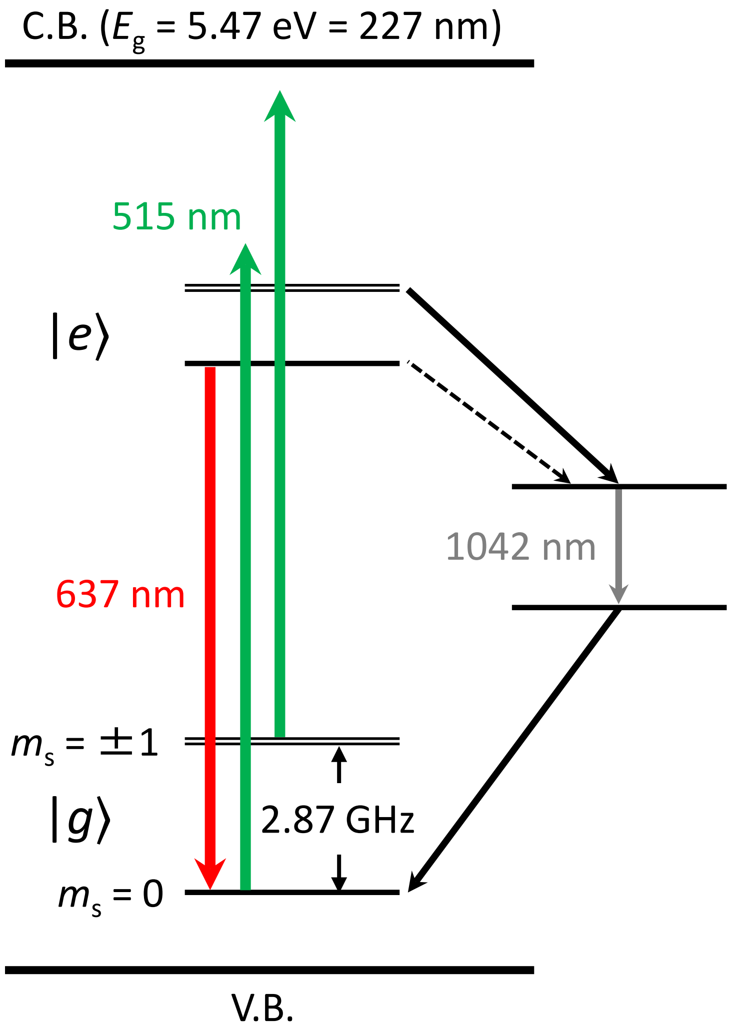
One photon at a time



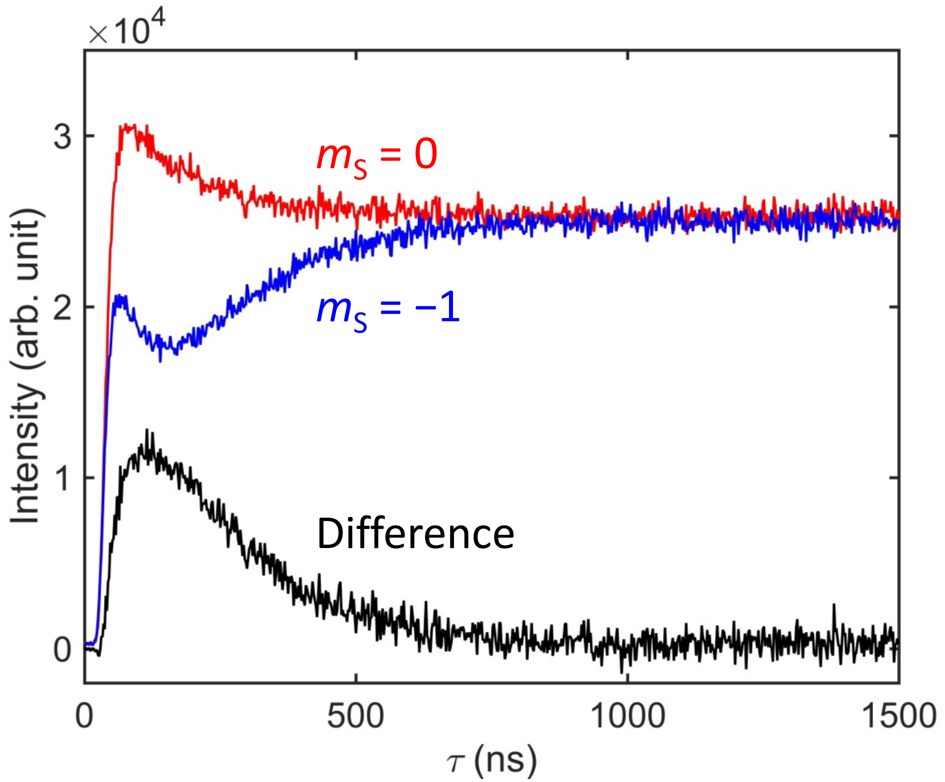
Non-radiative path



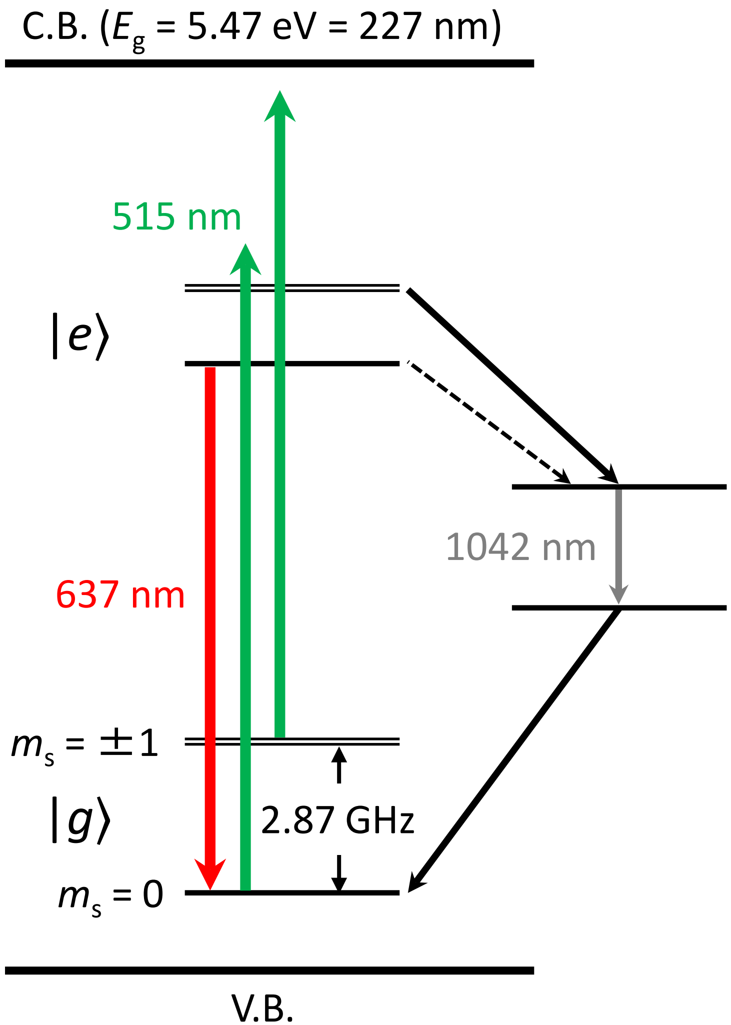
Time-resolved fluorescence

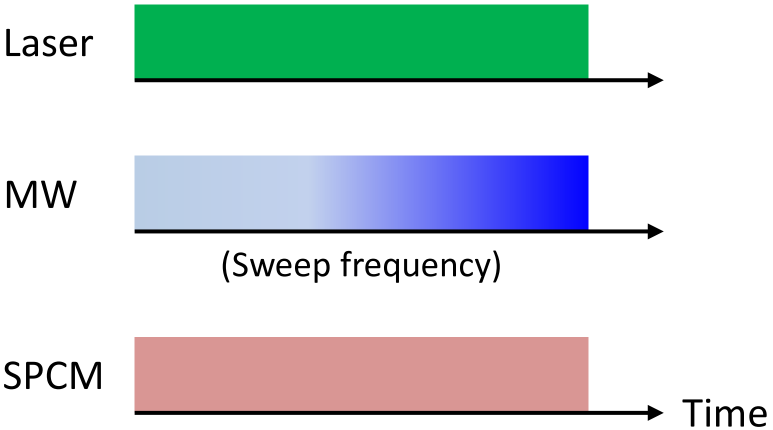


The NR channel provides a means to **read out** and **initialize** the NV spin

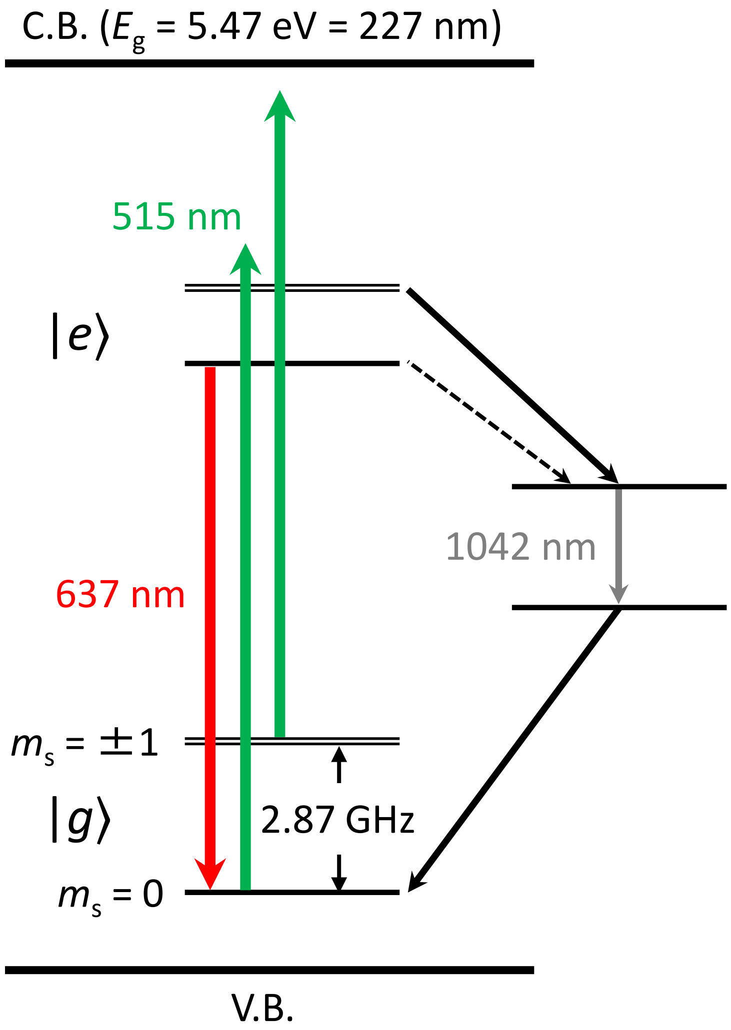


CW ODMR


C.B. (E_g = 5.47 eV = 227 nm)
515 nm
|e⟩
637 nm
1042 nm
m_s = ±1
|g⟩
2.87 GHz
m_s = 0
V.B.

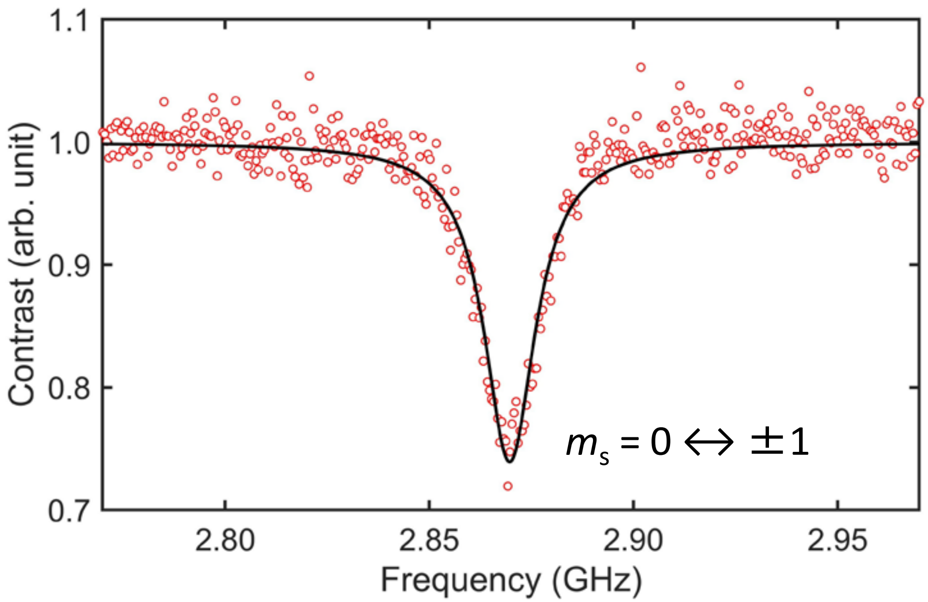

Laser
MW
(Sweep frequency)
SPCM
Time

CW ODMR at $B_0 = 0$

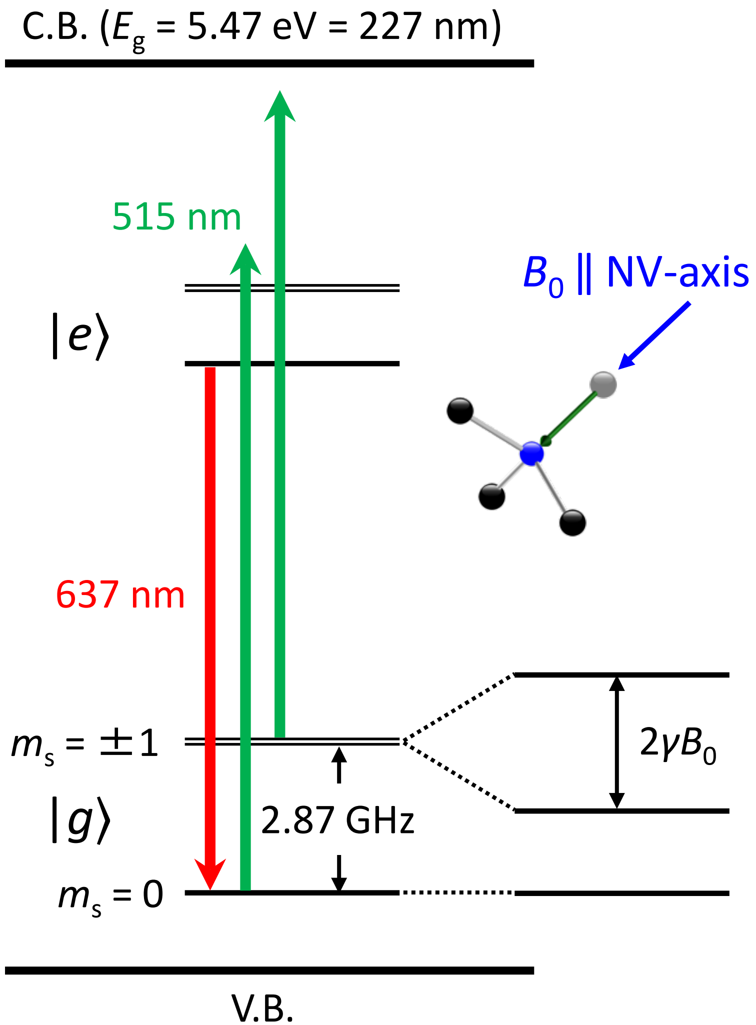


Zero-field splitting $H = DS_z^2$

D = 2.87 GHz

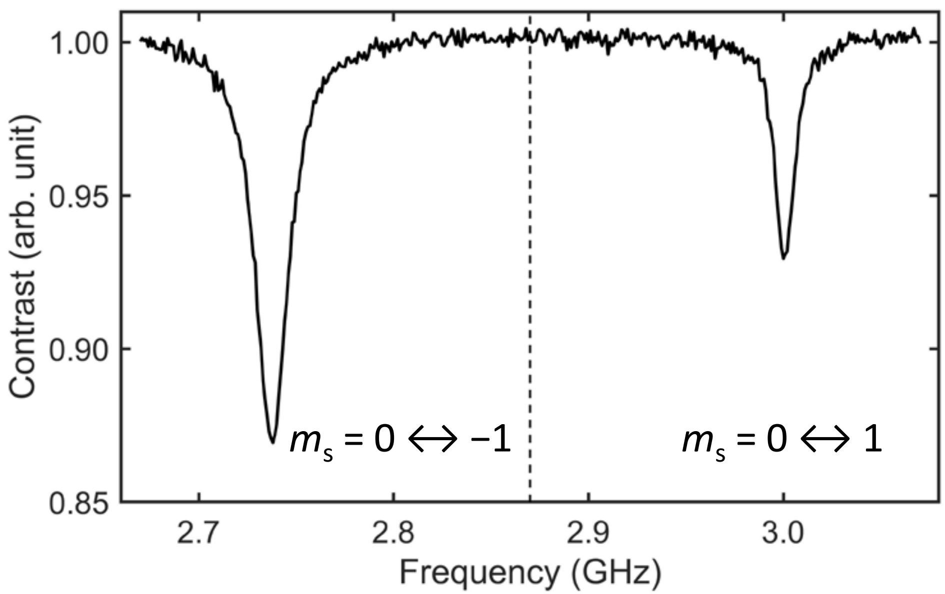


CW ODMR at $B_0 > 0$



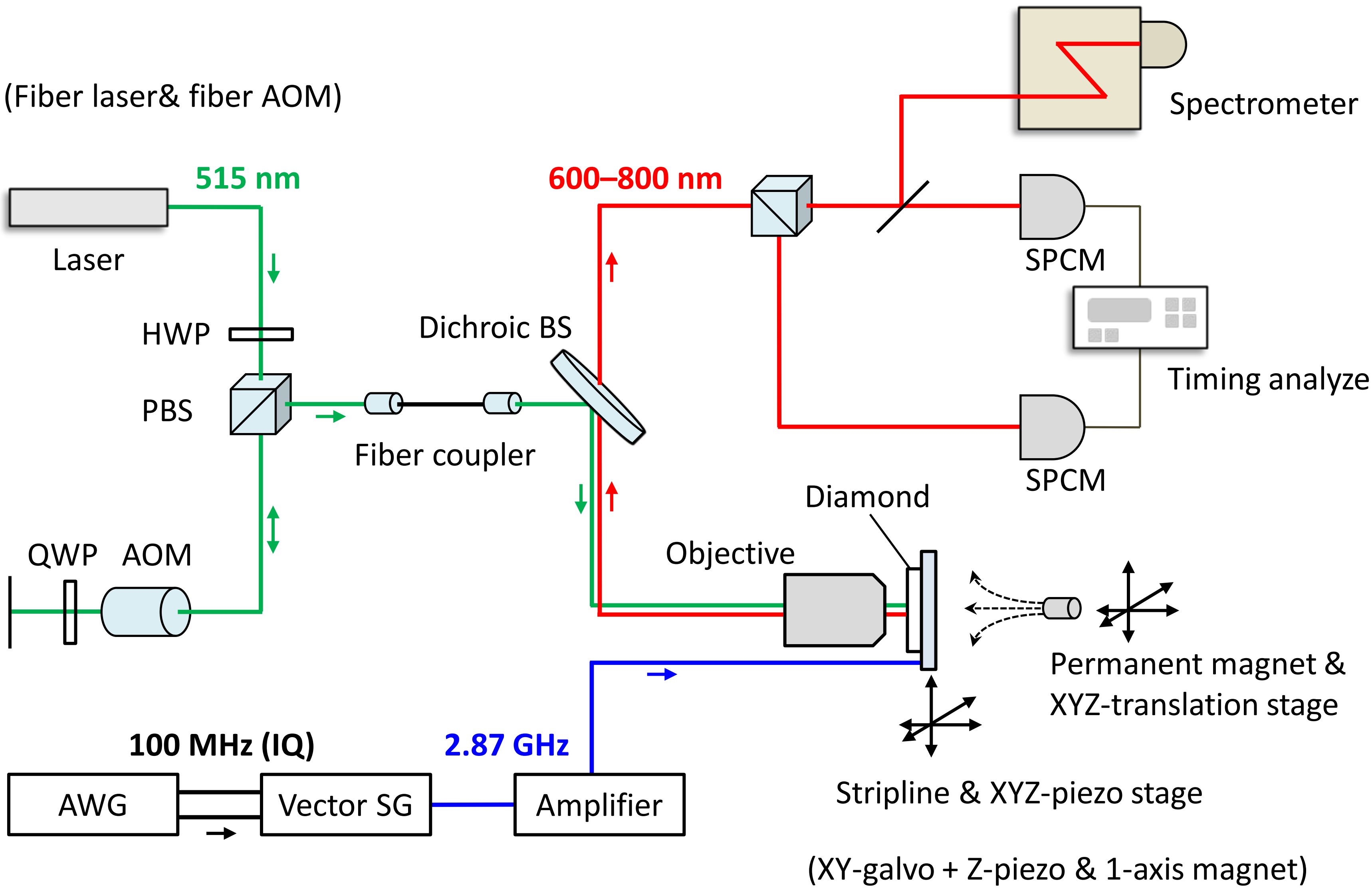
Zeeman $H = DS_z^2 + \gamma_e B_0 S_z$

γ_e = 28 MHz/mT

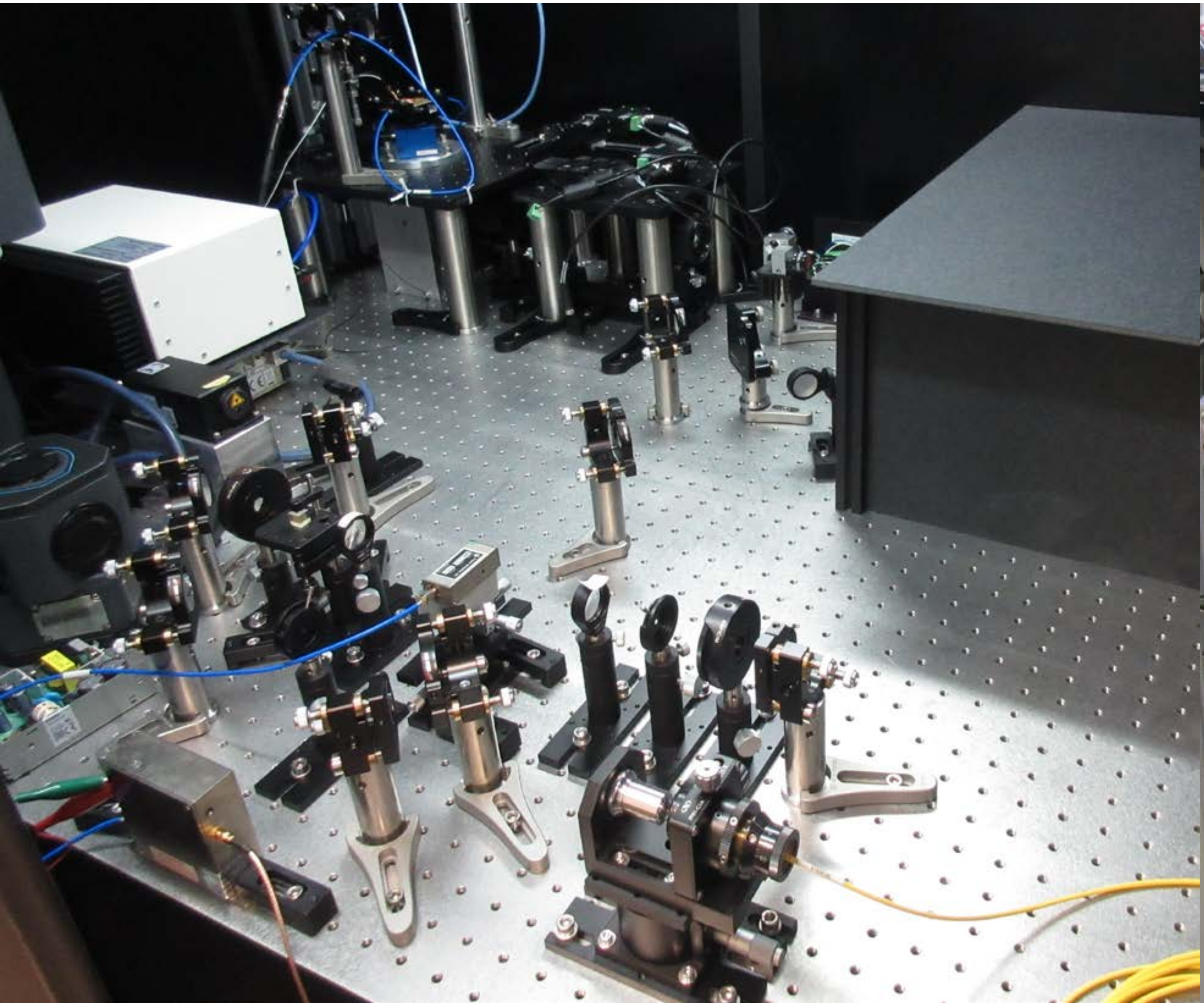


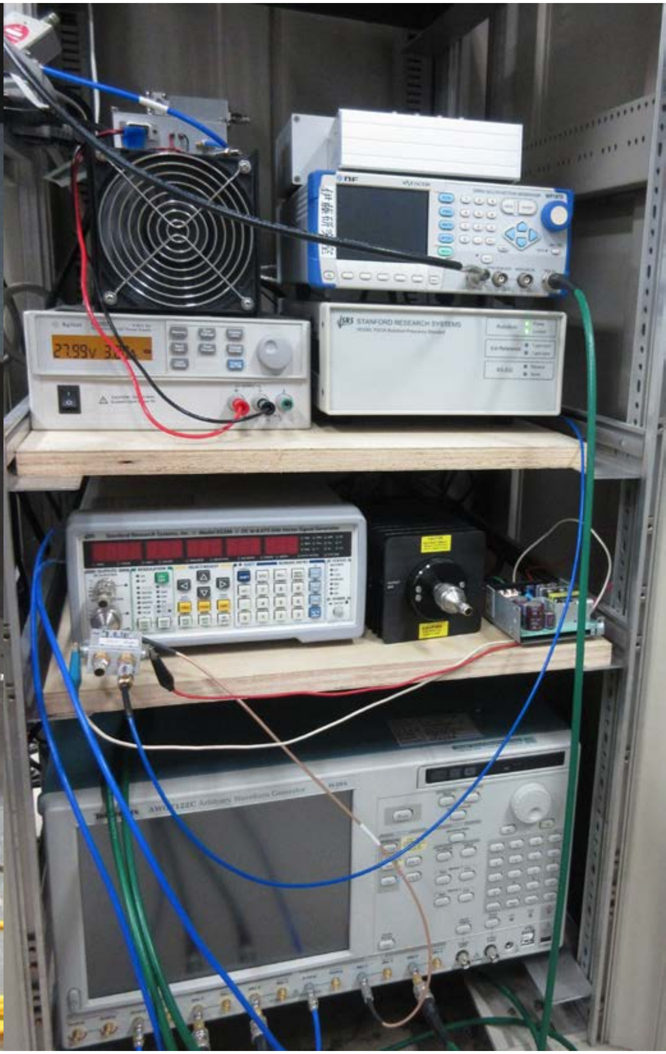
B_0 = 4.7 mT (2.87±0.132 GHz)

Experimental setup



Experimental setup





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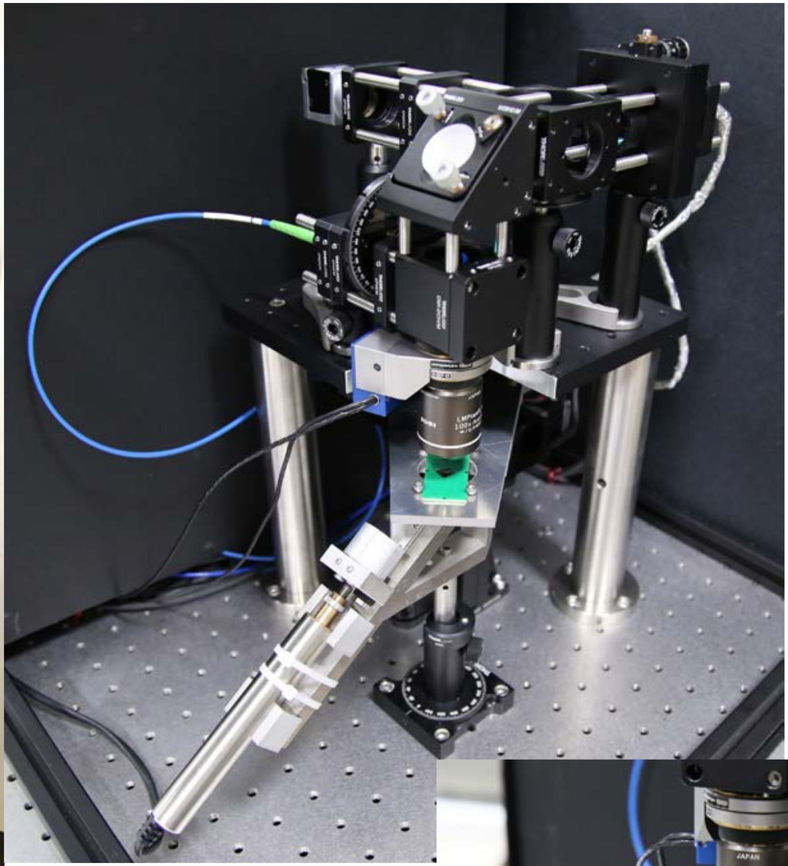
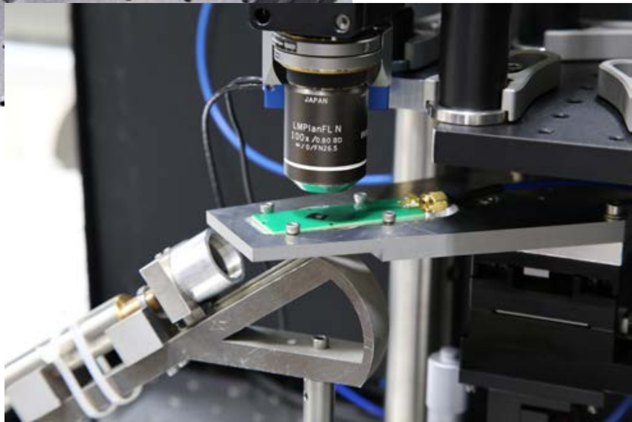
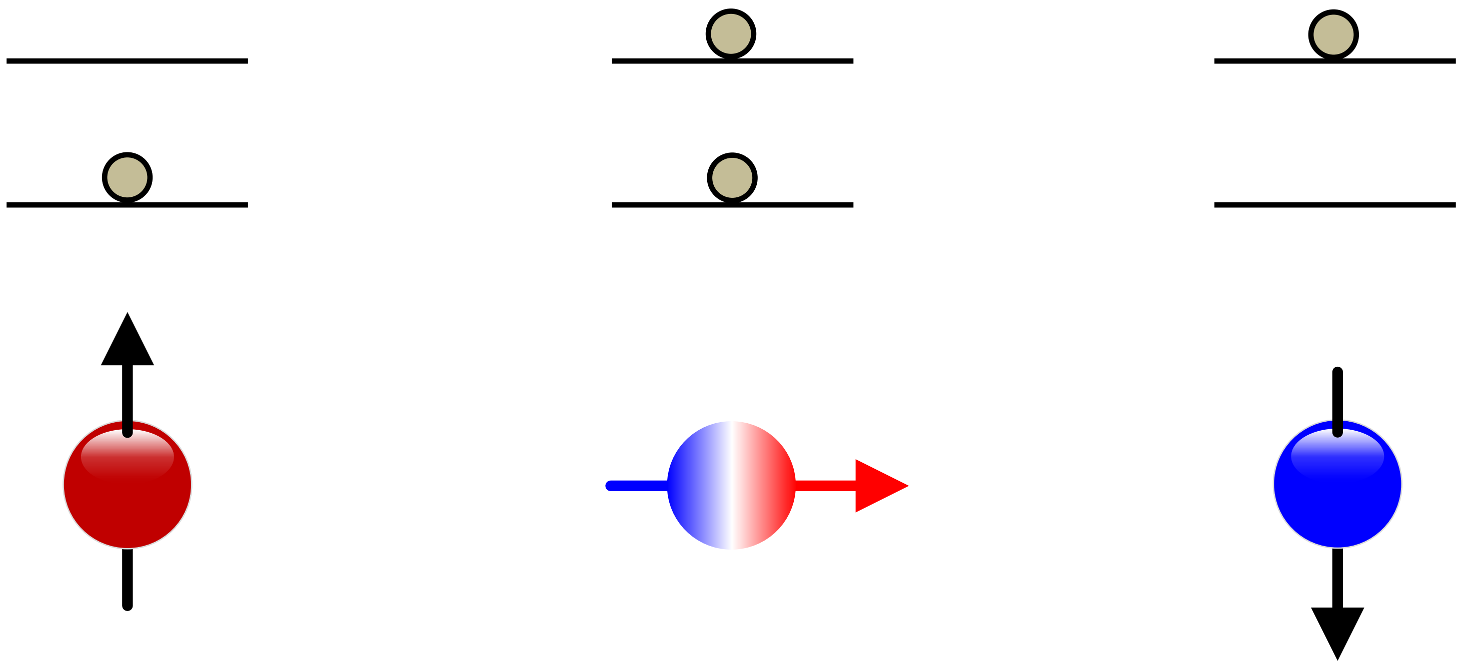




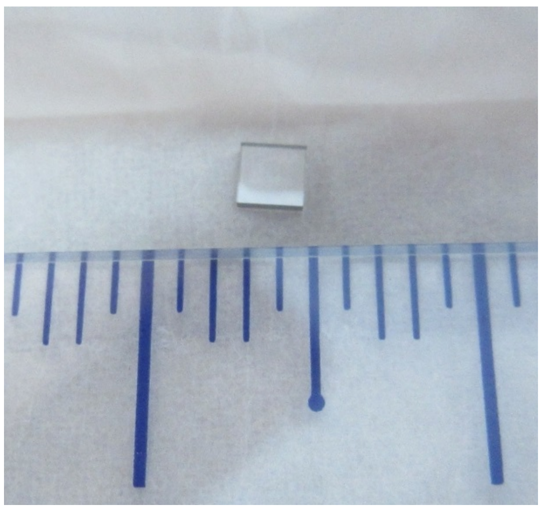
Qubit & coherence



$|"0"\rangle \equiv |m_s = 0\rangle$ $\quad$ $|\Psi\rangle = \alpha|0\rangle + \beta|1\rangle$ $\quad$ $|"1"\rangle \equiv |m_s = -1\rangle$

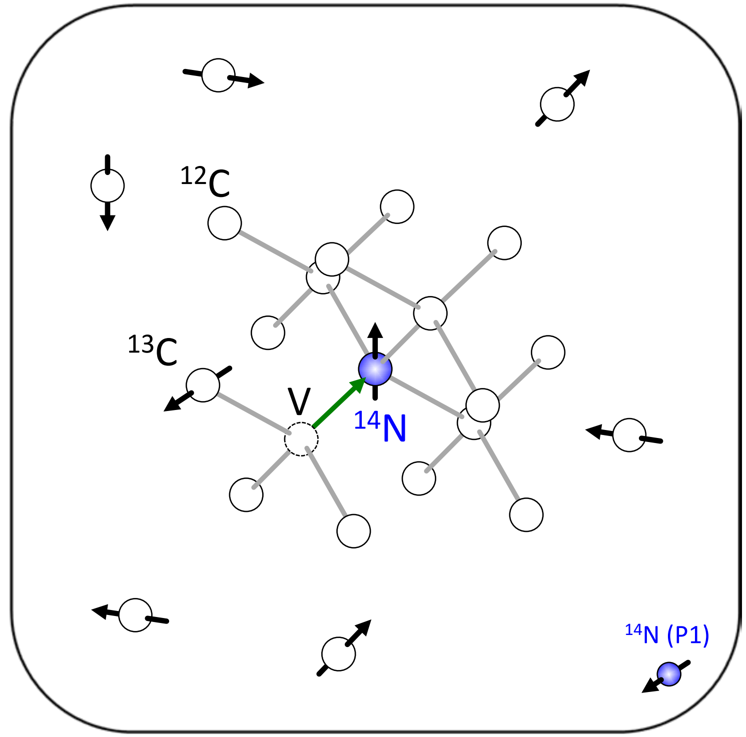
***T_2*: measure of how long a superposition state is preserved**

Magnetic environment



[N] < 5 ppb, [NV] < 0.03 ppb

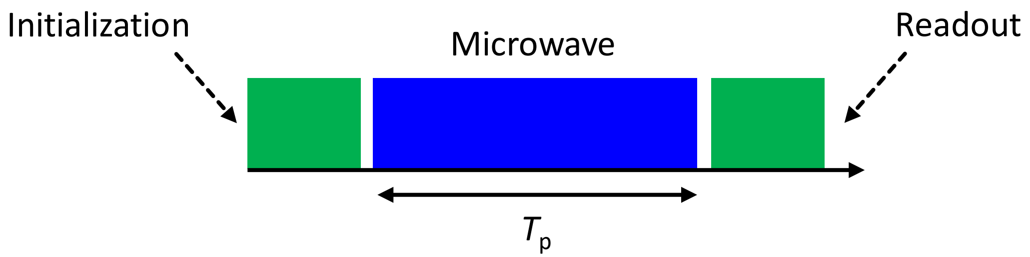
$\rho_N = 1.77 \times 10^{23}$ cm^{-3}

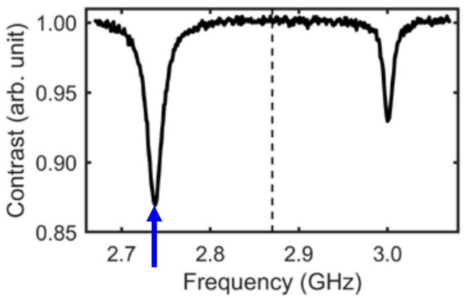


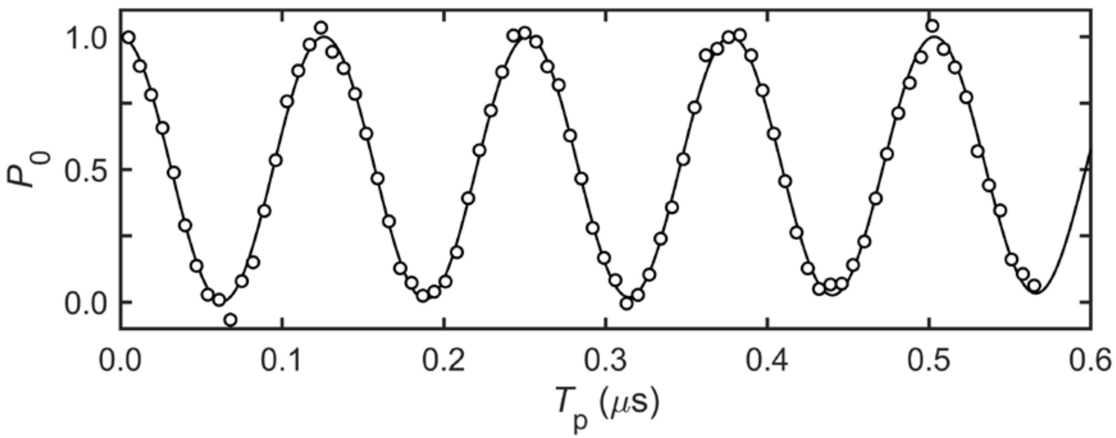
^{12}C (I = 0) : ^{13}C (I = ½) = 98.9% : 1.1%

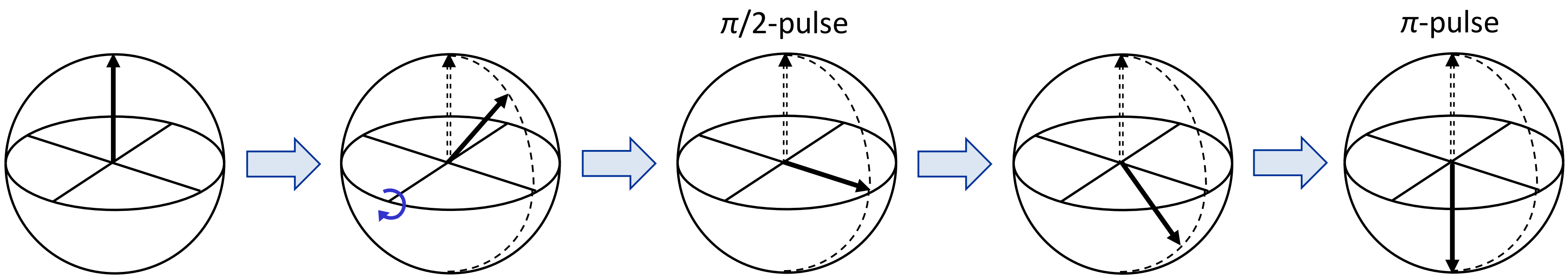
^{14}N (I = 1) : ^{15}N (I = ½) = 99.6% : 0.4%

Rabi oscillation

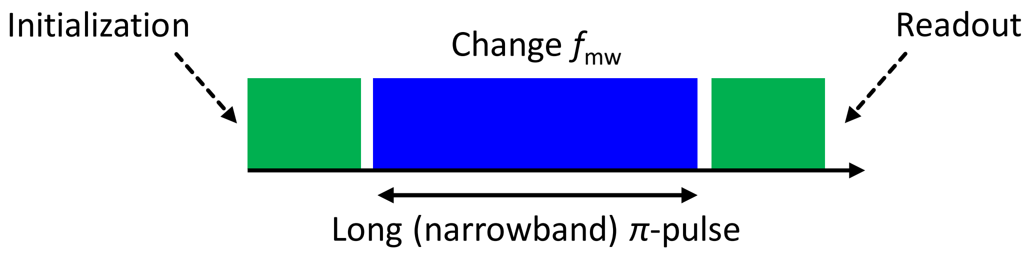


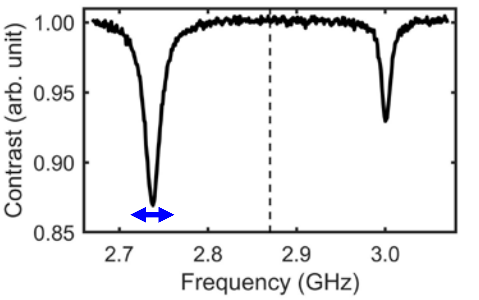


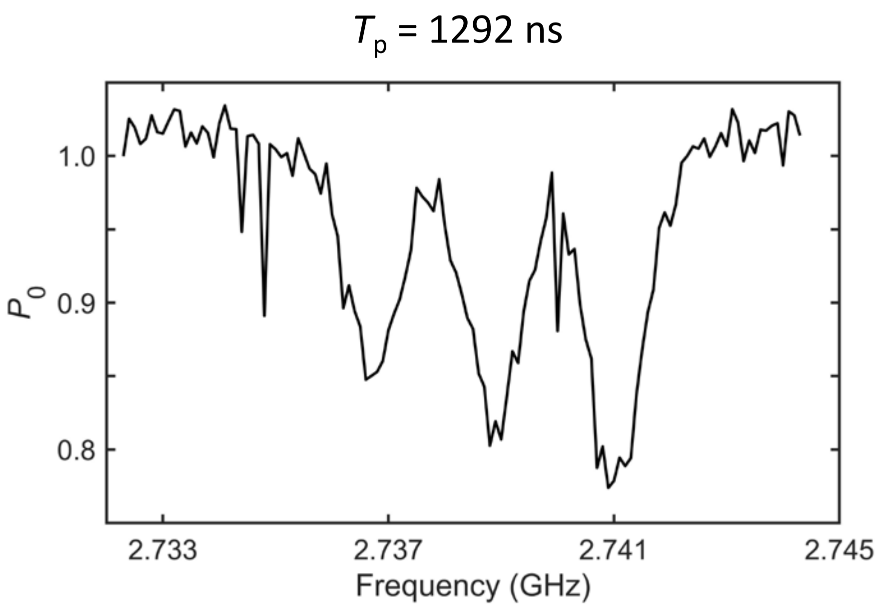


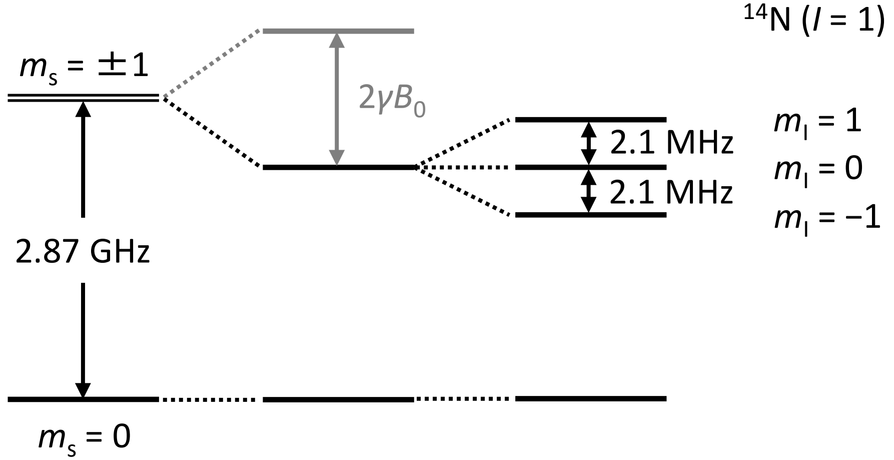


Pulsed ODMR



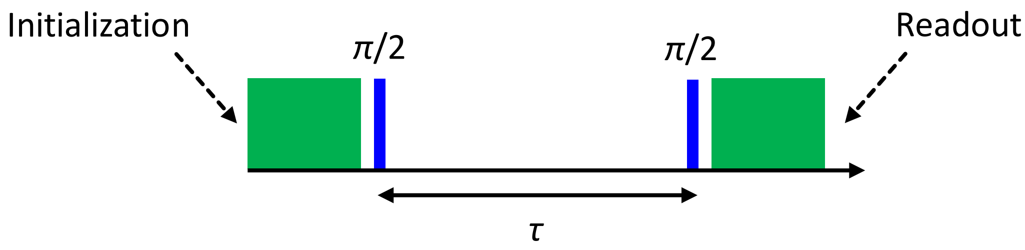


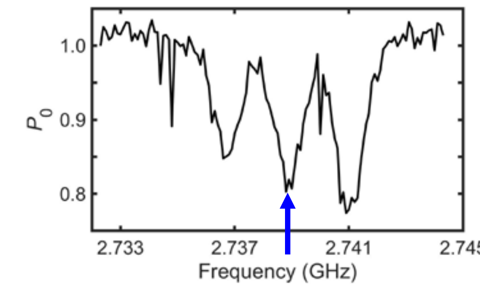


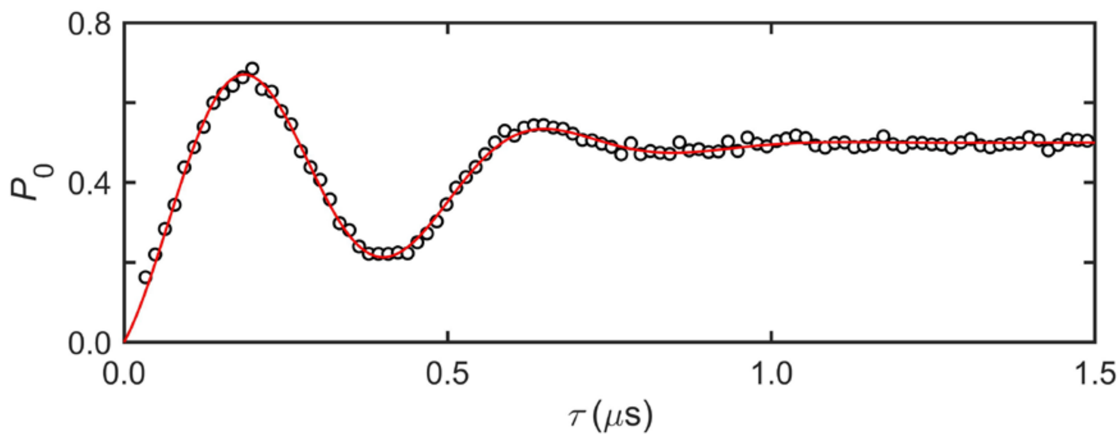


Hyperfine coupling with ^{14}N nucleus of the NV center itself

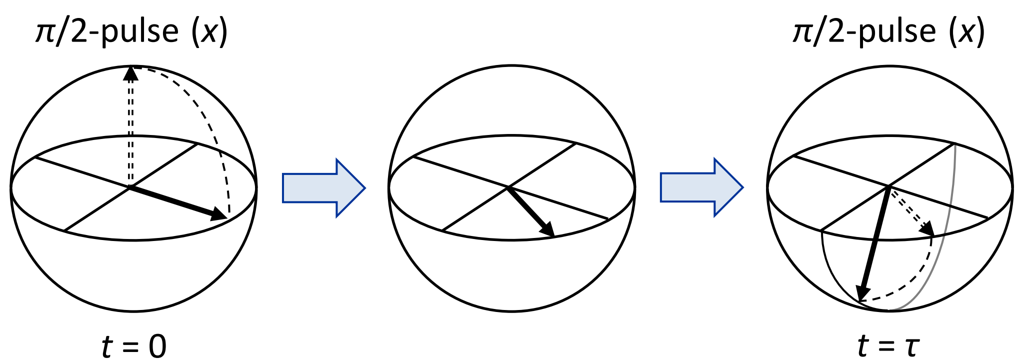
Ramsey interference



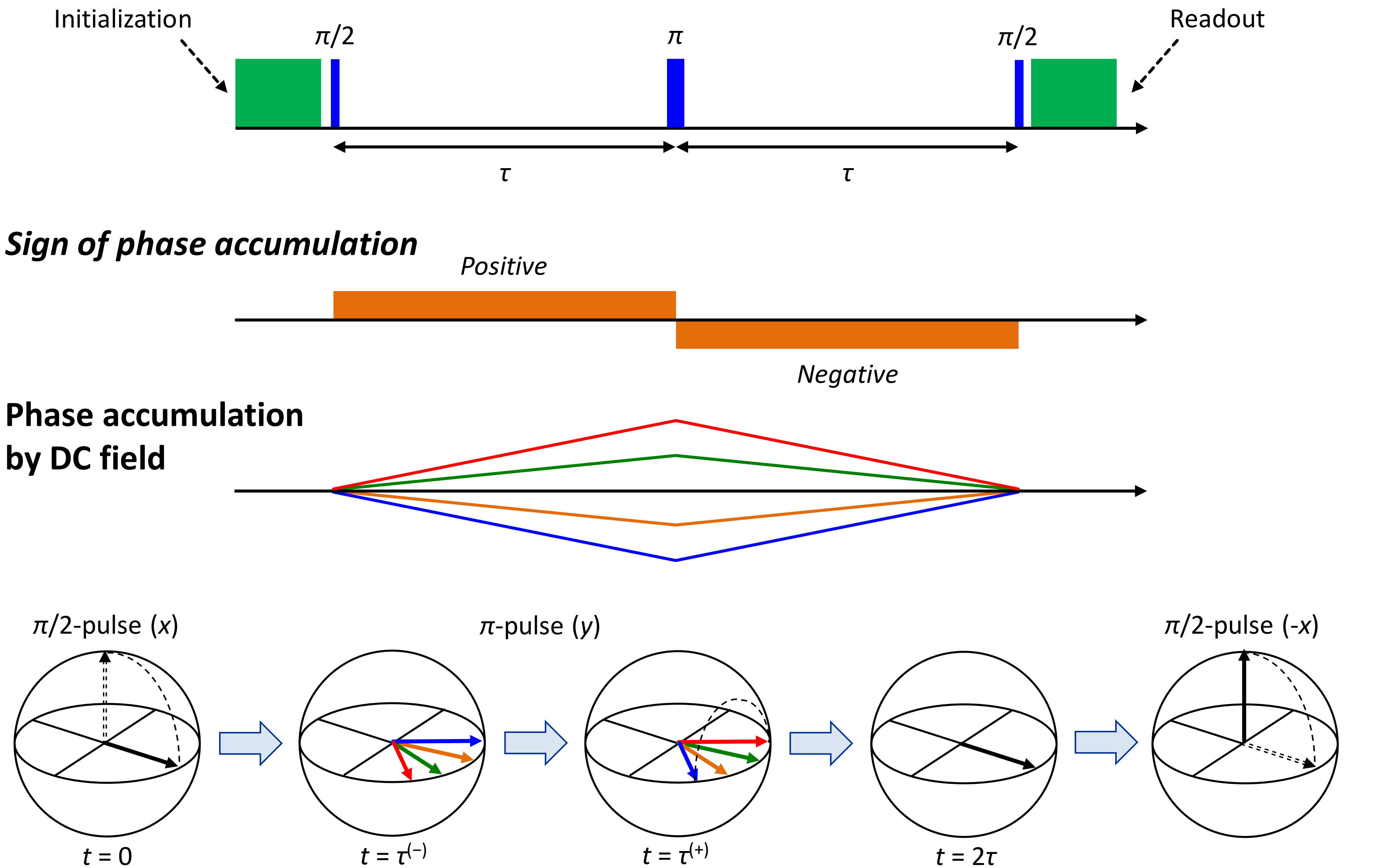




$T_{\pi/2}$ = 31.5 ns
f_{osc} = 2.1 MHz
T_2* = 0.50 μs

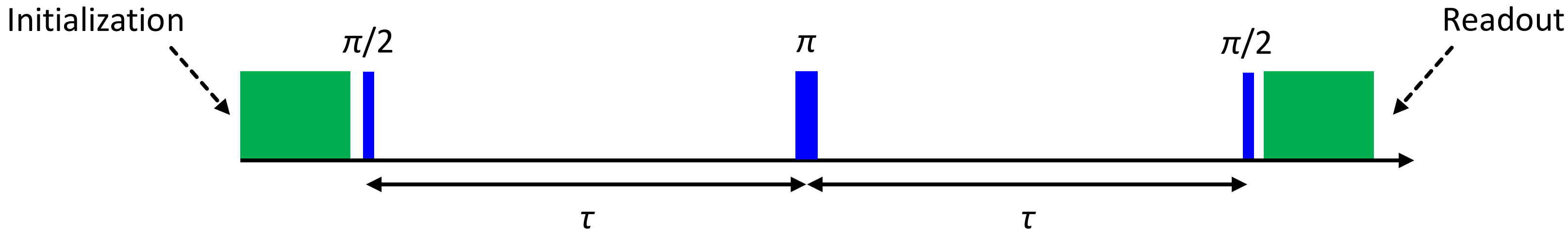


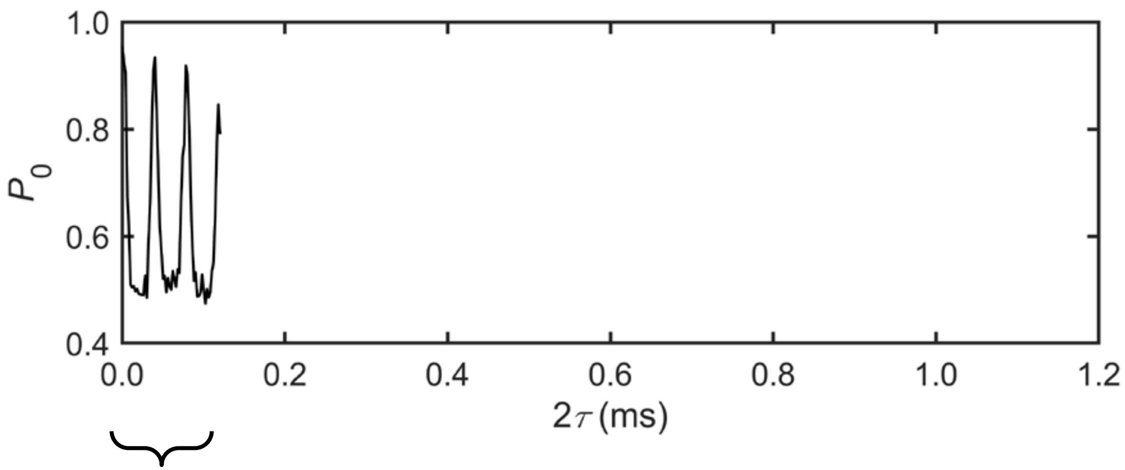
Spin echo



Initialization
π/2
π
π/2
Readout
τ
τ
Sign of phase accumulation
Positive
Negative
Phase accumulation
by DC field
π/2-pulse (x)
π-pulse (y)
π/2-pulse (-x)
t = 0
t = τ(−)
t = τ(+)
t = 2τ

Spin echo

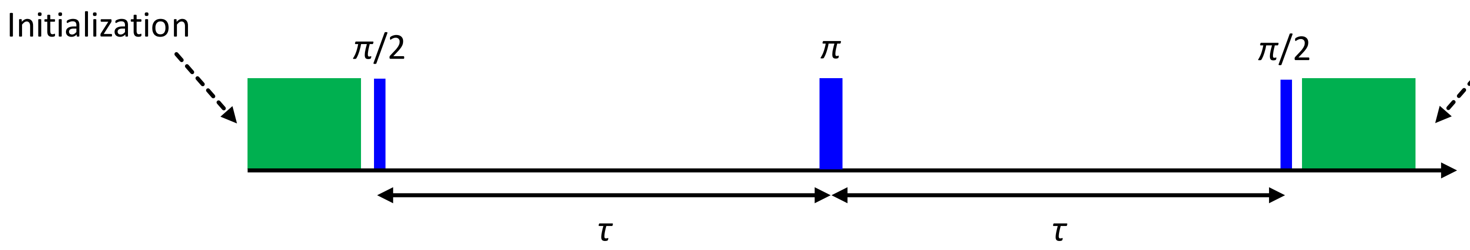


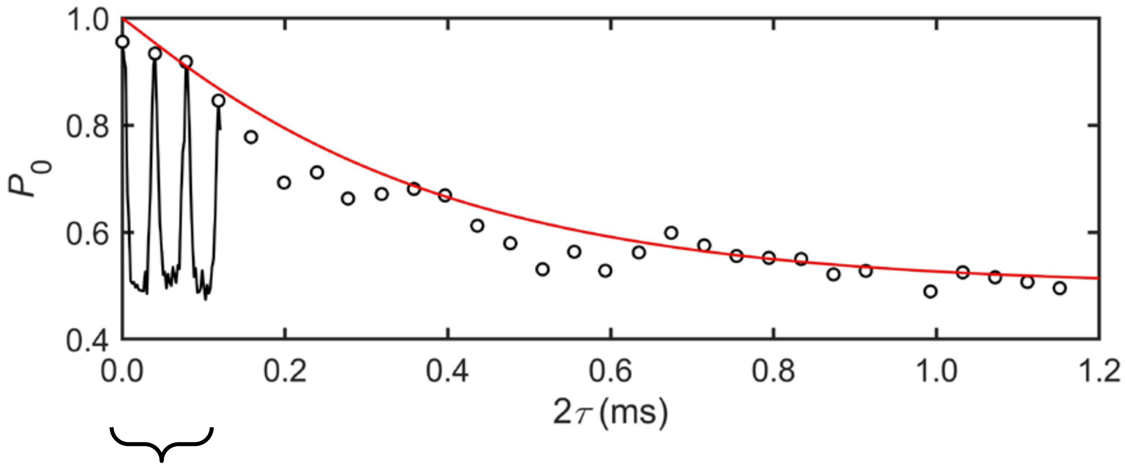


"Collapse and revival"

- Interaction with the nuclear spin bath ([^{13}C] = 1.1%)
- $f_c = \gamma_C B_0$ = 10.705 kHz/mT x 4.7 mT = 50.3 kHz
- Revival at $2n\tau = 2n/f_c$ = 41.8 x n μs

Spin echo



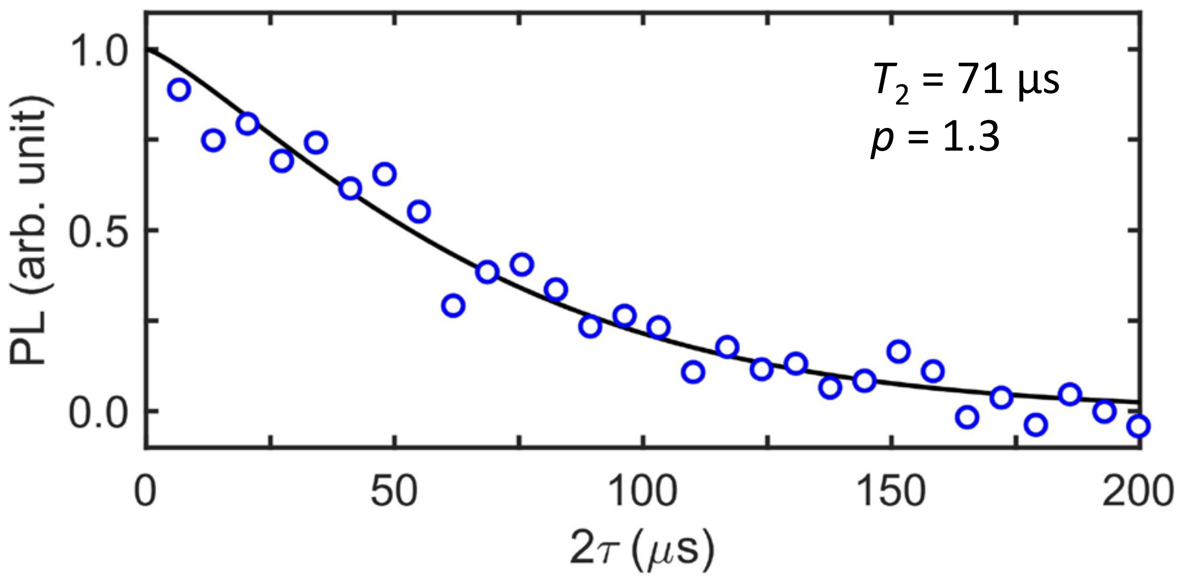


T_2 = 364 μs

"Collapse and revival"

- Interaction with the nuclear spin bath ($[^{13}C]$ = 1.1%)
- $f_c = \gamma_C B_0$ = 10.705 kHz/mT x 4.7 mT = 50.3 kHz
- Revival at $2n\tau = 2n/f_c$ = 41.8 x n μs

Coherence time



Stretched exponential decay

$$\exp\left[-\left(\frac{2\tau}{T_2}\right)^p\right]$$

CVD growth of shallow single NV centers

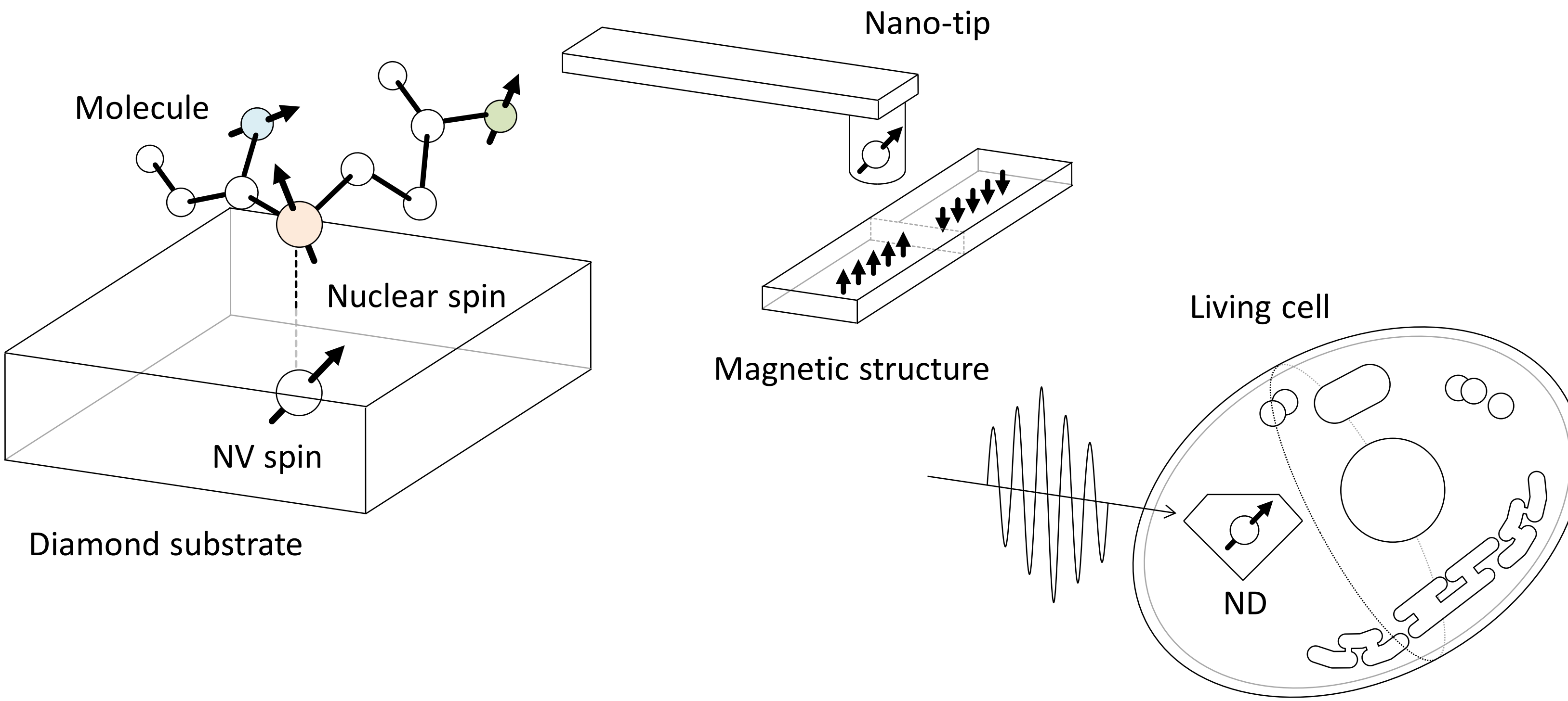
- Hydrogen-terminated
- ~5 nm from the surface
- [^{12}C] = 99.999%

Outline

- **Basics of NV centers in diamond**
  - Structure
  - Optical properties
  - Spin properties

- **Quantum sensing**
  - Basics
  - Correlation spectroscopy and detection of nuclear spins
  - Ultrahigh resolution sensing
  - Determination of the position of a single nuclear spin

Quantum sensing

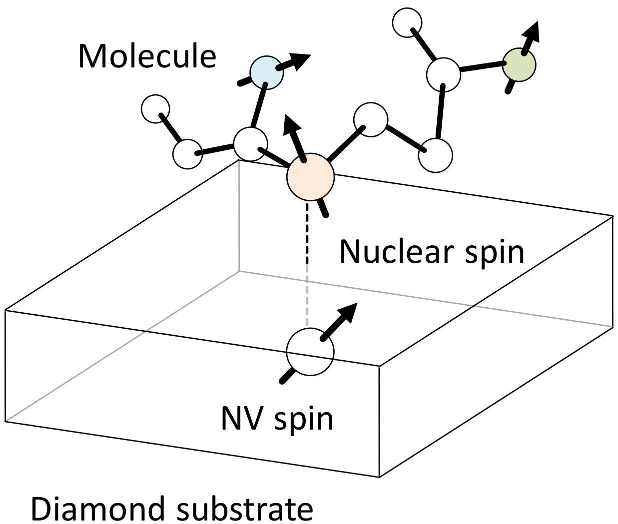


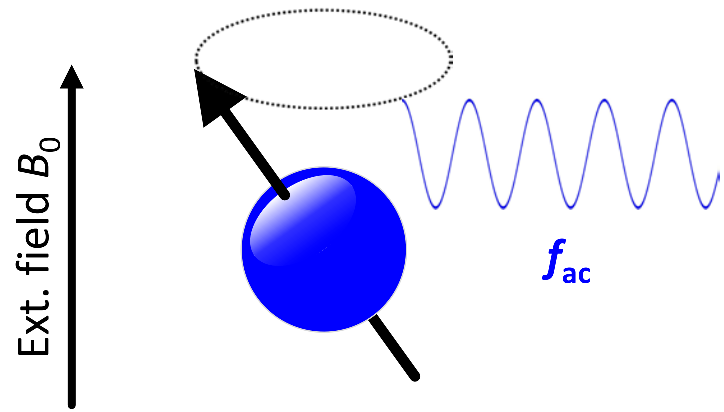
- Room T. operation
- High spatial resolution
- Nondestructive
- Various modalities

⇨

- Nano MRI
- Probe for CM systems
- Biology

Nuclear spin sensing





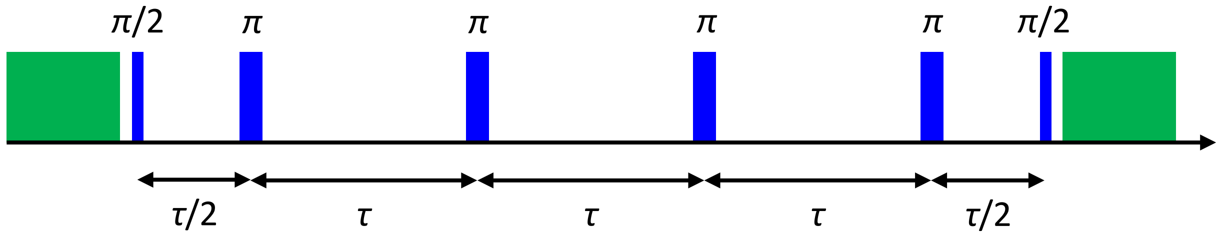
Nuclear spins **precess** at f_{ac} = a few kHz–MHz under B_0

⇨ **Weak AC magnetic field** on the NV spin (11 nT@d_{NV} = 5 nm)

⇨ Detect using **quantum coherence**

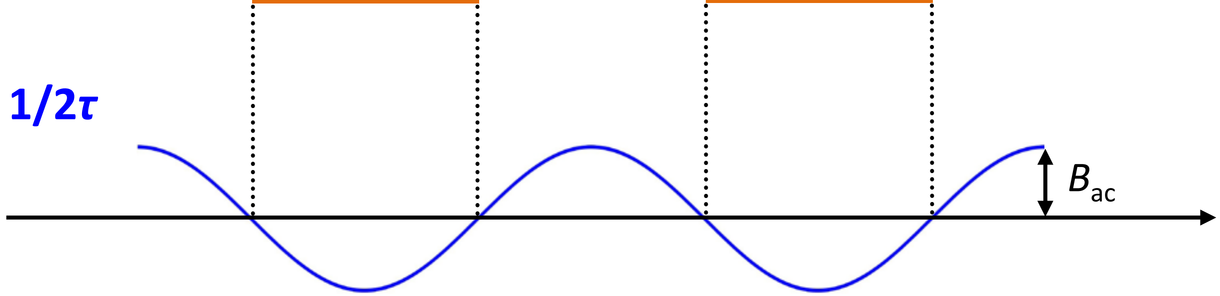
AC magnetometry

CP (*N* = 4)



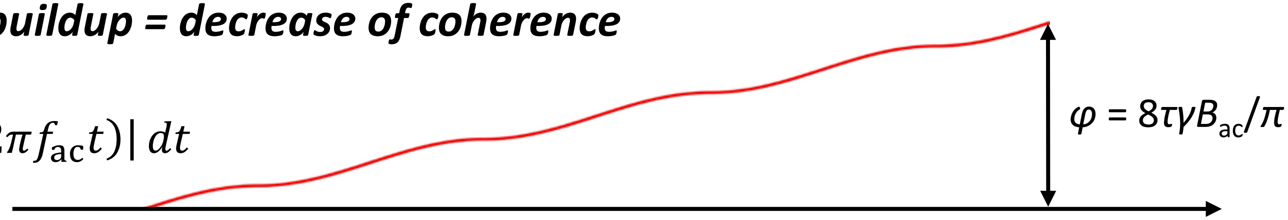
Sign of phase accumulation

AC field at f_{ac} = 1/2τ



Sensor phase buildup = decrease of coherence

$$\gamma B_{\mathrm{ac}} \int_0^t |\cos(2\pi f_{\mathrm{ac}} t)|\, dt$$



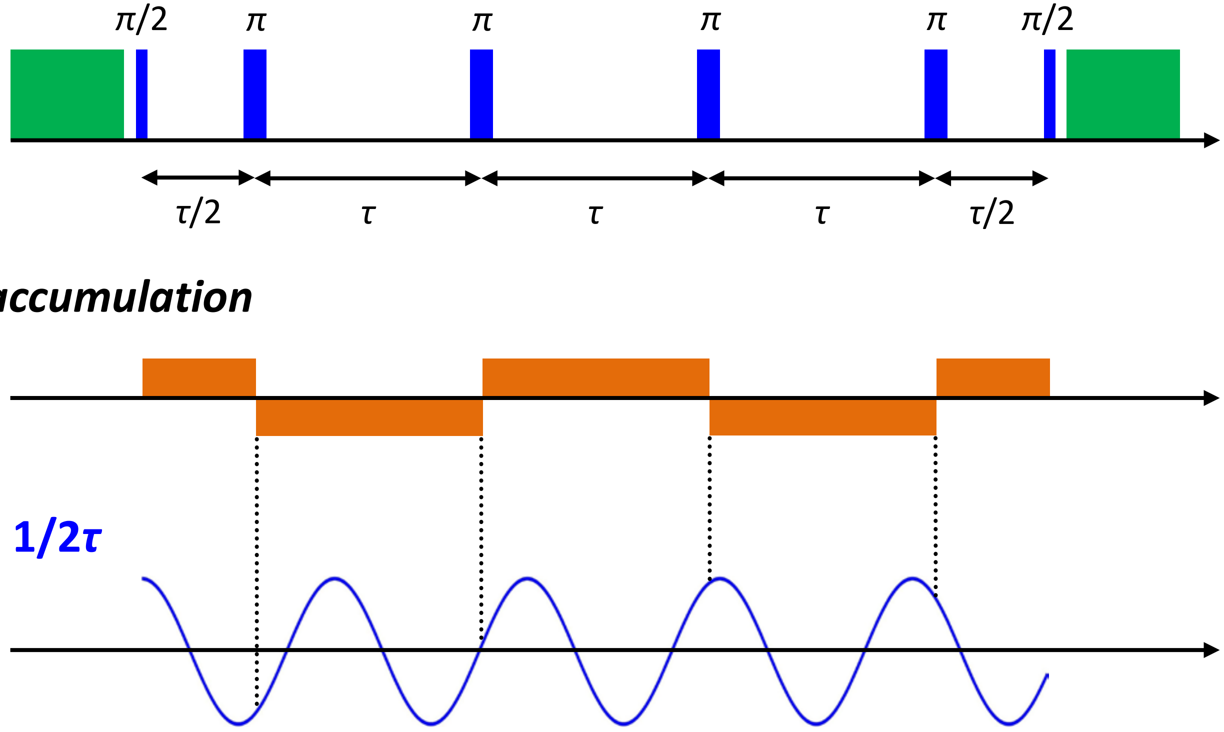
Rev. Mod. Phys. **89**, 035002 (2017) Degen *et al.*

AC magnetometry

CP (N = 4)

Sign of phase accumulation

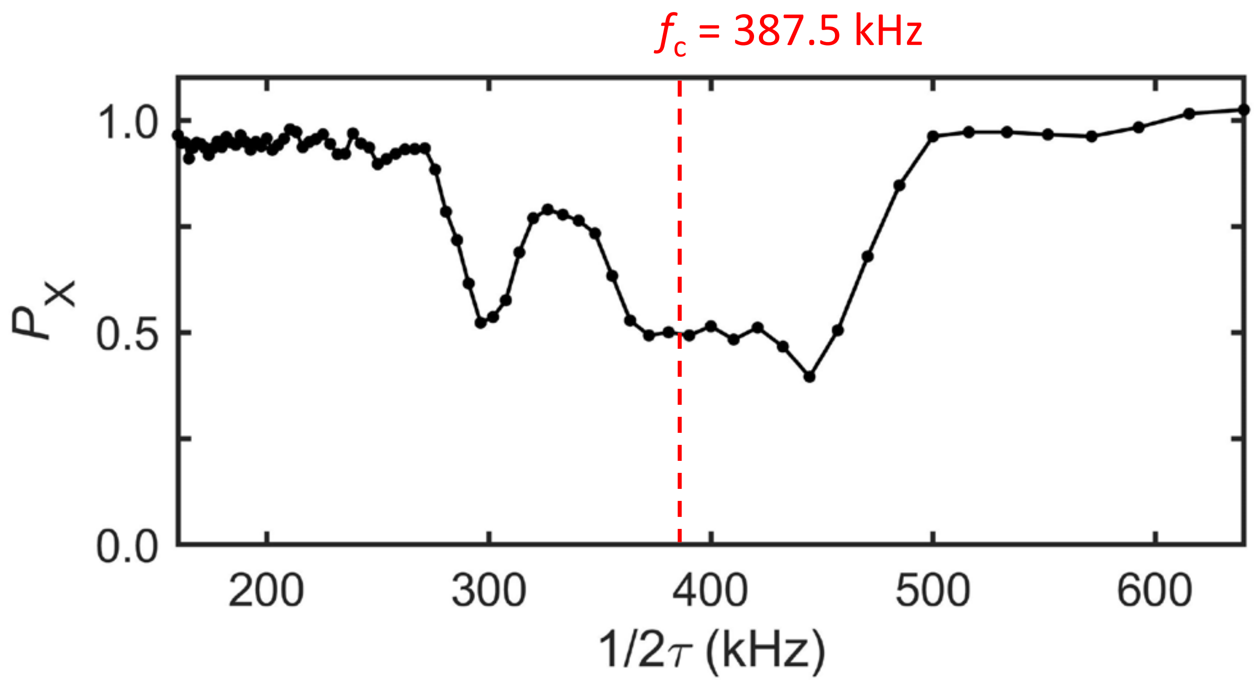
AC field at $f_{ac} \neq 1/2\tau$

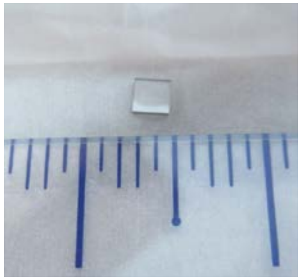


Multiple oscillations between π-pulses average out the sensor phase

Rev. Mod. Phys. **89**, 035002 (2017) Degen *et al.*

Nuclear spin sensing

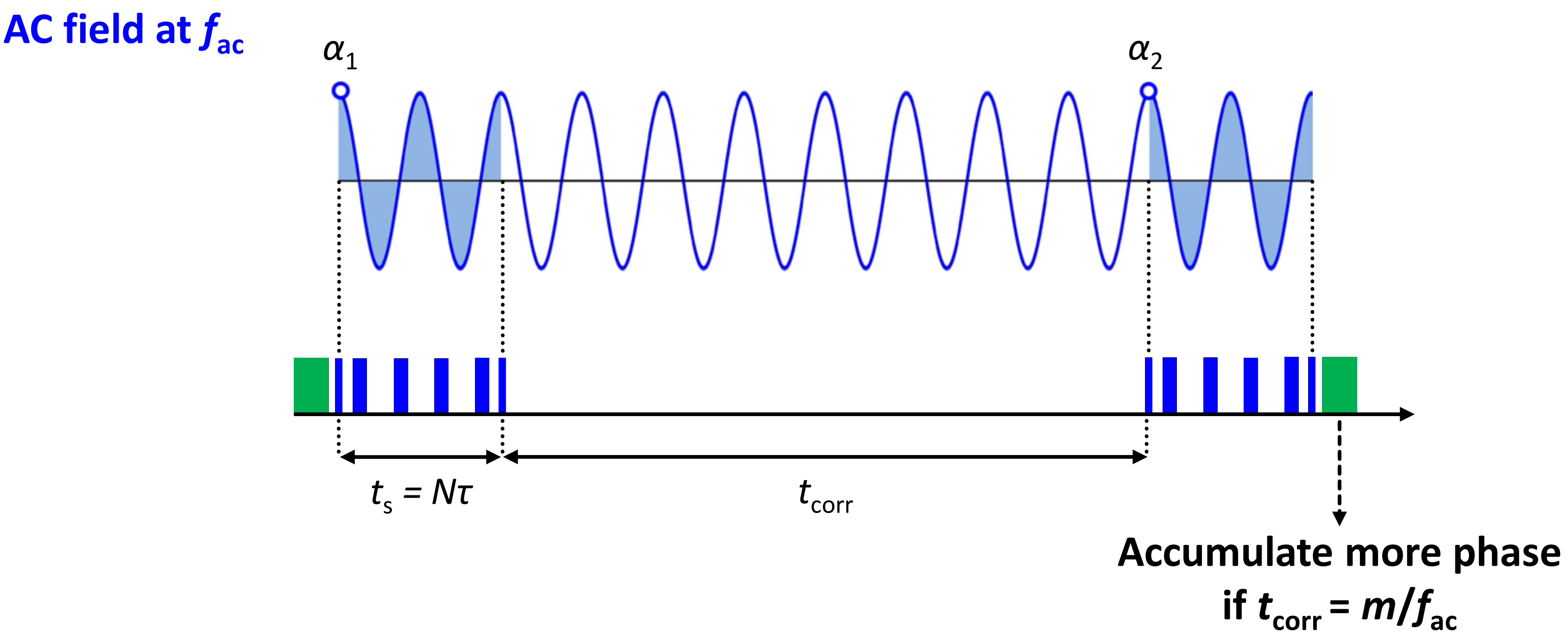




- Single NV center in a natural abundant diamond ($[^{13}C]$ = 1.1%, d_{NV} ~ 50 μm)
- **Sweep τ & repeat** ($f = 1/2\tau$, $\Delta\tau$ = 31.3 ns, N = 16)
- $f_c = \gamma_C B_0$ = 10.705 kHz/mT x 36.2 mT

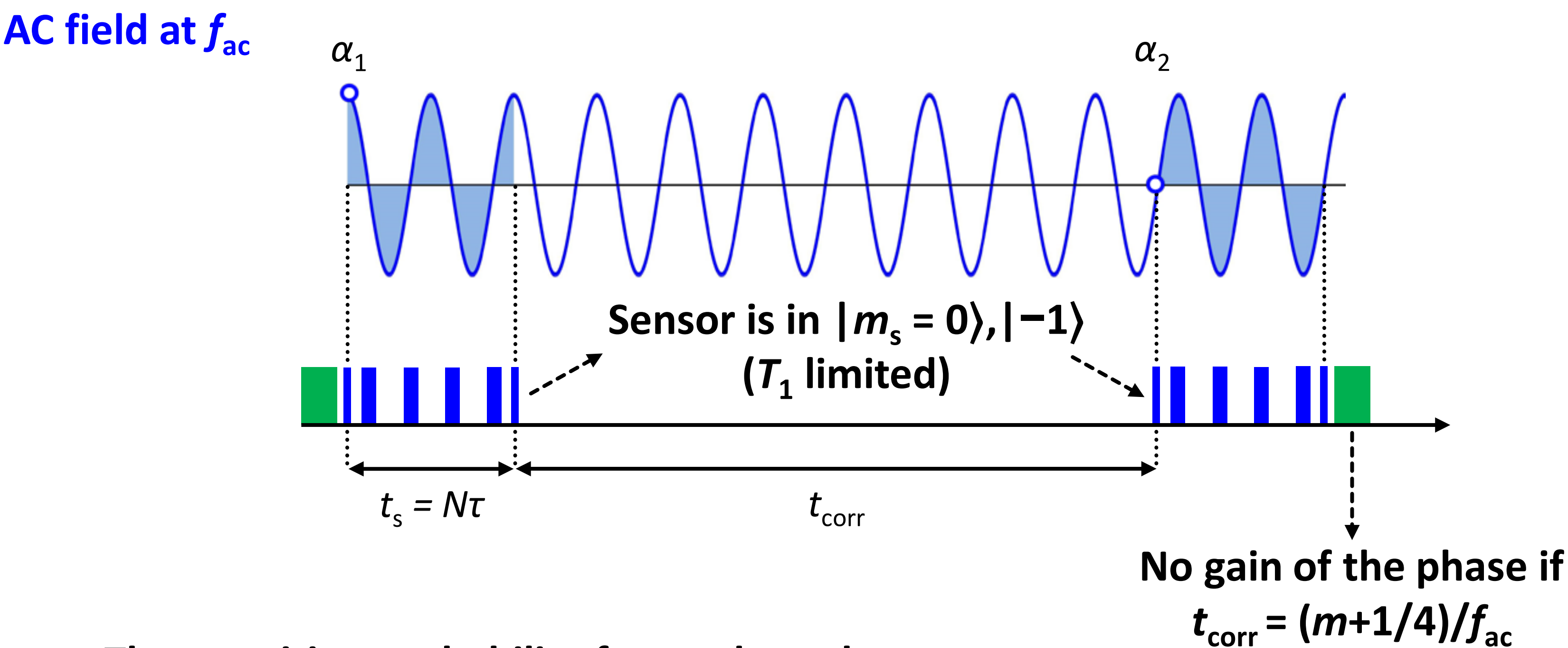
Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

Correlation spectroscopy



Nature Commun. **4**, 1651 (2013) Laraoui *et al.*
Phys. Rev. Appl. **4**, 024004 (2015) Kong *et al.*
Nature Commun. **6**, 8527 (2015) Staudacher *et al.*
Phys. Rev. Lett. **116**, 197601 (2016) Boss *et al.*

Correlation spectroscopy



The transition probability for random phases

$$p(t_1) \approx \frac{1}{2}\left\{1 - \frac{1}{2}\left(\frac{\gamma B_{ac} t_s}{\pi}\right)^2 \cos(2\pi f_{ac} t_{corr})\right\}$$

Nature Commun. **4**, 1651 (2013) Laraoui *et al.*
Phys. Rev. Appl. **4**, 024004 (2015) Kong *et al.*
Nature Commun. **6**, 8527 (2015) Staudacher *et al.*
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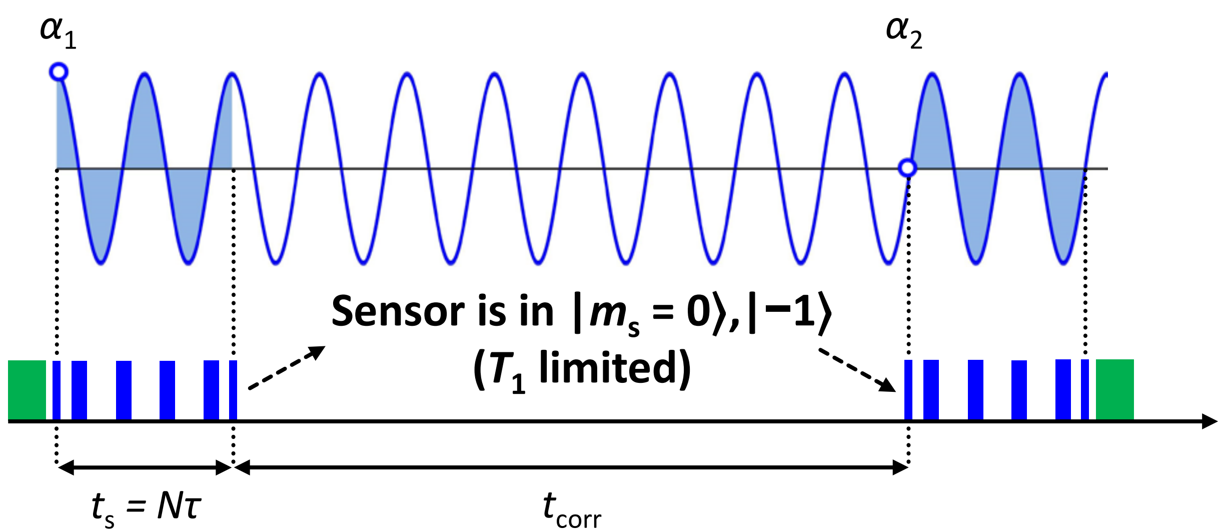
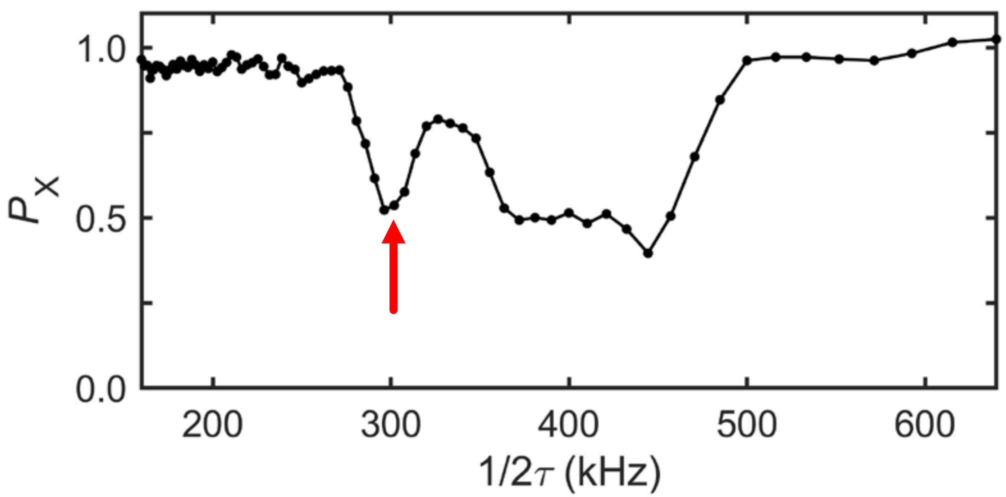
Correlation spectroscopy

AC field at f_{ac}



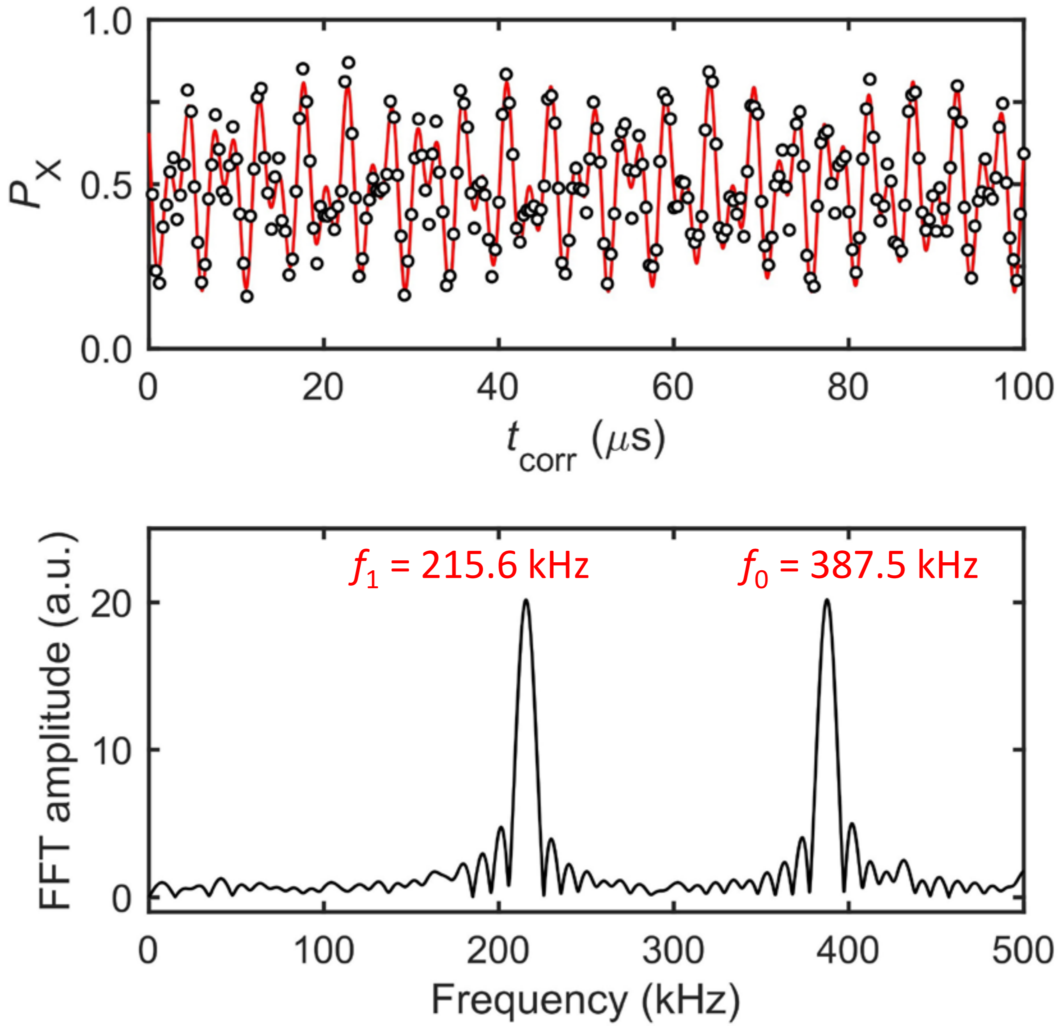


Where to look at?

- $f_t = 1/2\tau = 301.6$ kHz
- $\tau = 1.7875$ µs

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

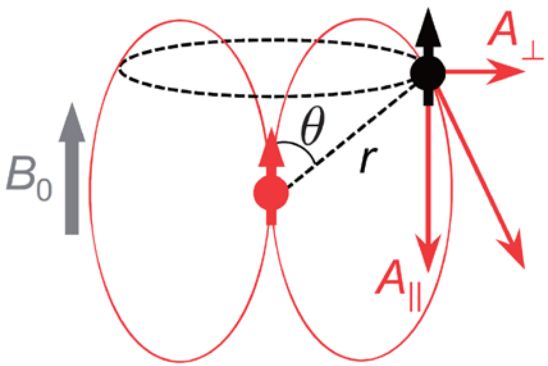
Correlation spectroscopy of a nucleus



No decay up to 100 μs

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

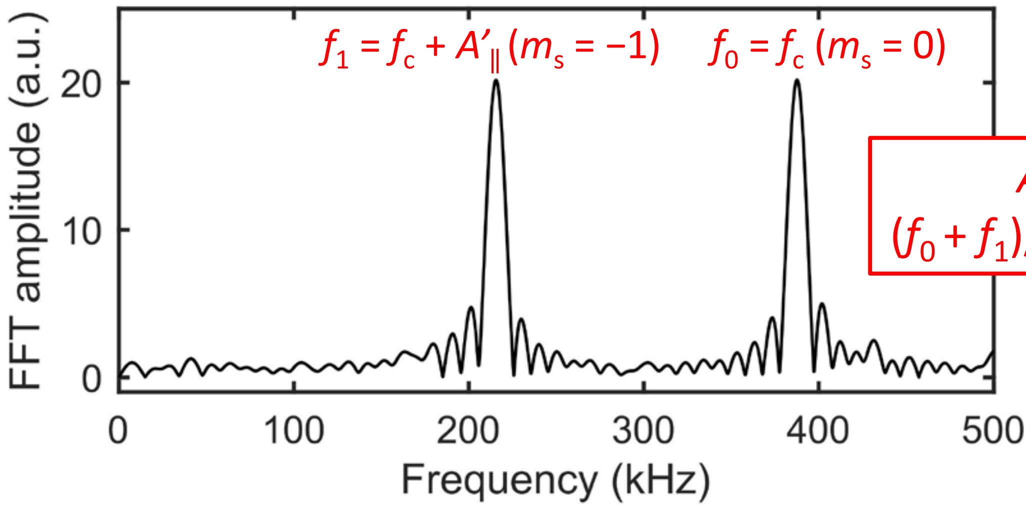
Correlation spectroscopy of a nucleus



Hamiltonian of NV–^{13}C coupled system

$$H = f_c I_z + |m_s = -1\rangle\langle -1|(A_\parallel I_z + A_\perp I_x)$$

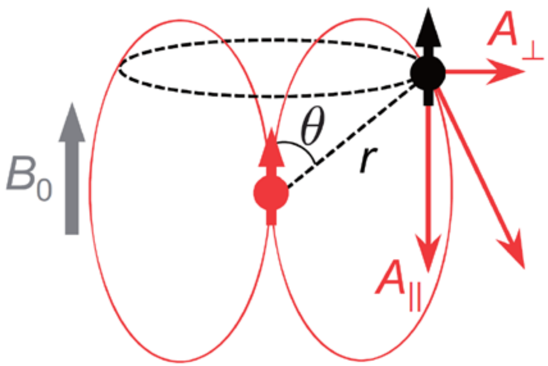
→ No hyperfine field when $|m_s = 0\rangle$



$A'_\parallel = -171.9$ kHz
$(f_0 + f_1)/2 = 301.55$ kHz → dip

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

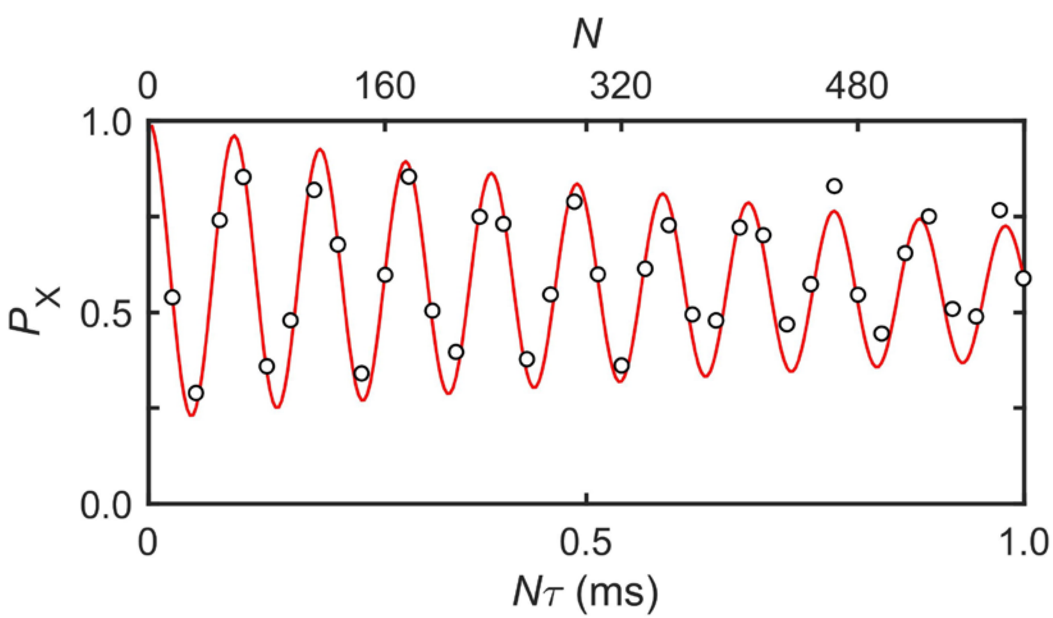
Coherent control of a nuclear spin



Hamiltonian of NV–^{13}C coupled system

$$H = f_c I_z + |m_s = -1\rangle\langle -1|(A_\parallel I_z + A_\perp I_x)$$

→ The single ^{13}C *n*-spin rotates about the $A_\perp$ axis

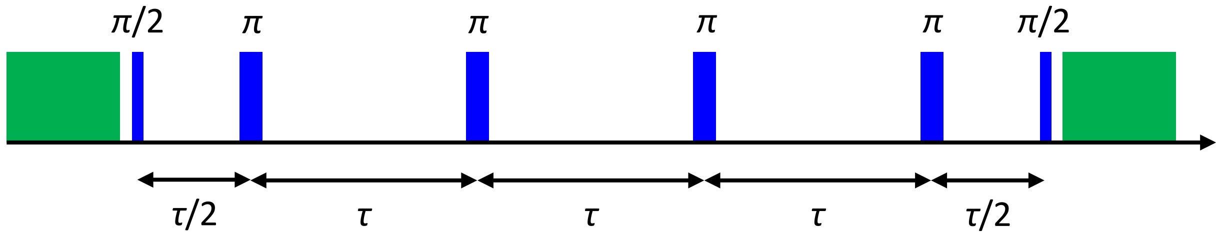


$$\tau \approx f_c + \frac{A_\parallel}{2}$$

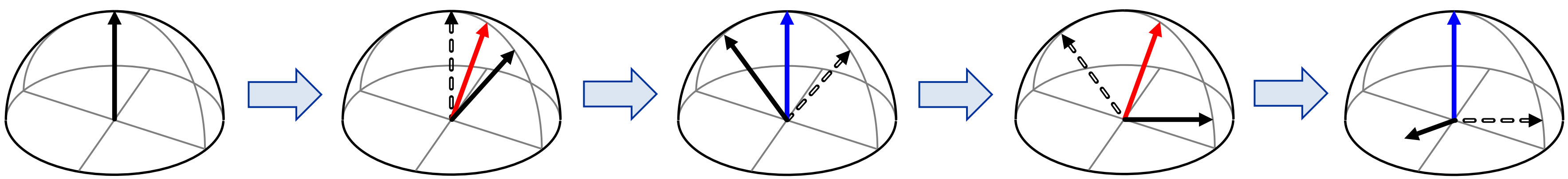
Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

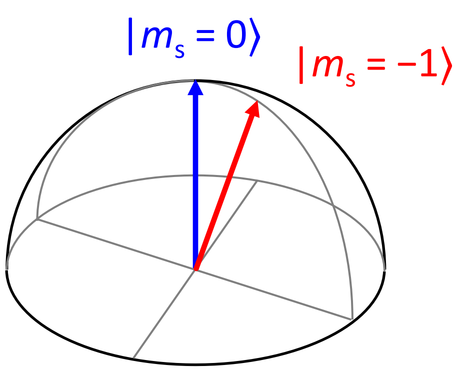
Conditional rotation of a nuclear spin

CP (*N* = 4)



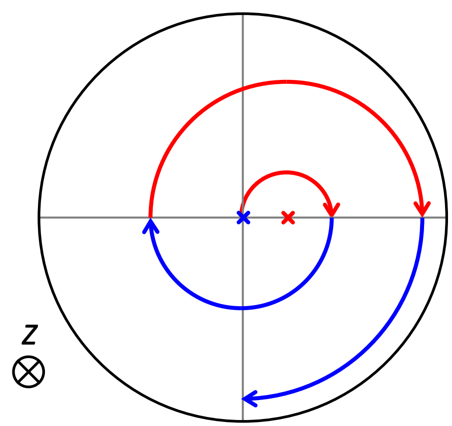
Evolution of *n*-spin vector



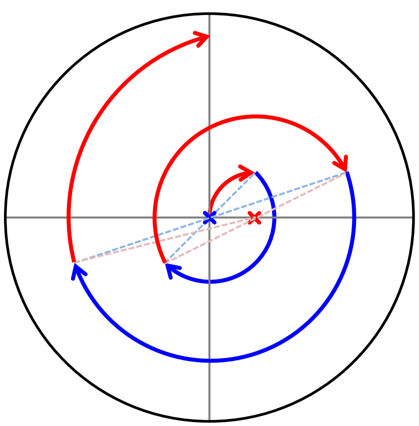


Q-axis of *n*-spin

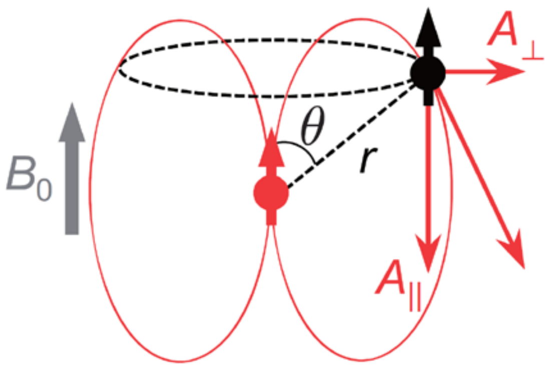
Start from |m_s = 0⟩



Start from |m_s = −1⟩



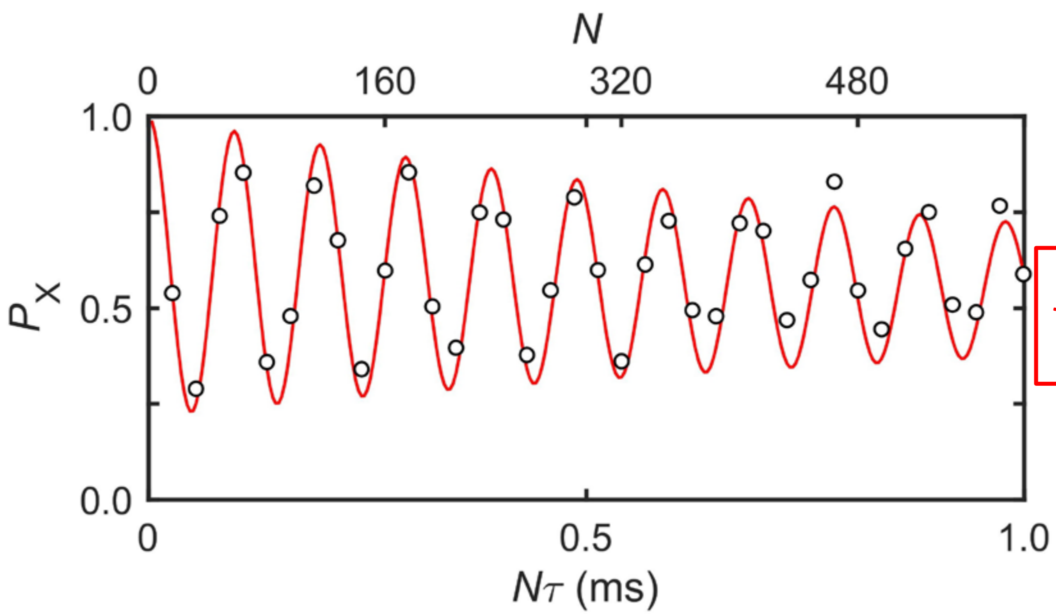
Coherent control of a nuclear spin



Transition probability of the NV spin

$$P_X = 1 - \frac{1}{2}(1 - \underbrace{\boldsymbol{n}_0 \cdot \boldsymbol{n}_{-1}}_{-1}) \sin^2 \frac{N\phi_{cp}}{2}$$

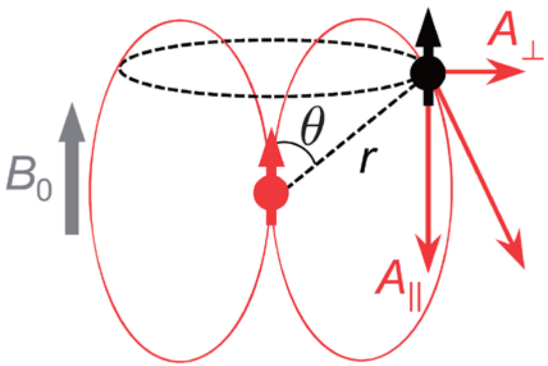
Phys. Rev. Lett. **109**, 137602 (2012) Taminiau *et al.*



f_{cp} = 10.2 kHz ≈ $A'_\perp/2$
P_X < 0.5 → single

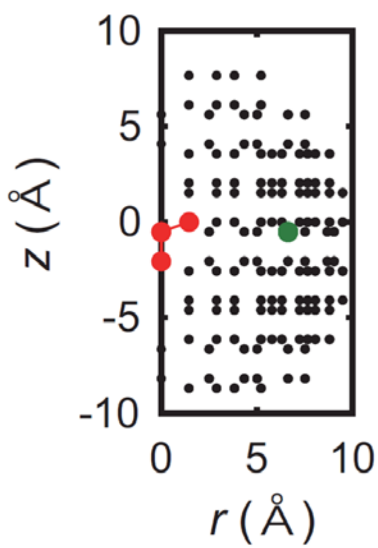
Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

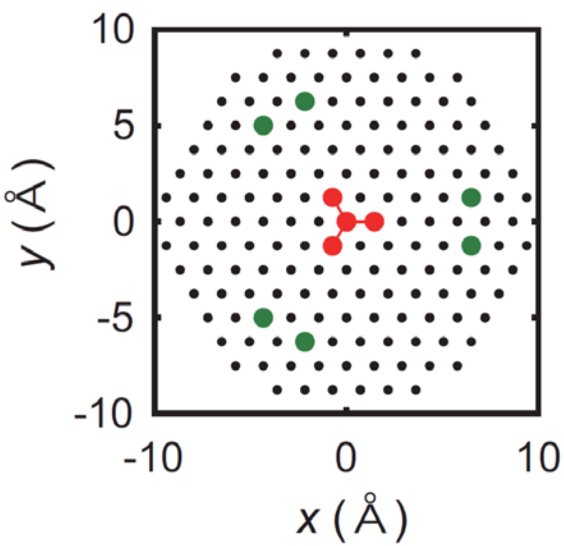
Determination of hf constants



Magnetic dipole int. + contact hf int.

$$A_\parallel \propto \frac{3\cos^2\theta - 1}{r^3} \qquad A_\perp \propto \frac{3\cos\theta\sin\theta}{r^3}$$





(r, θ) = (6.84 Å, 94.8°)

$A_\parallel$ = −173.1 kHz
$A_\perp$ = 22.3 kHz

$A_\parallel$ = −175.1±2.1kHz
$A_\perp$ = 21.9±0.2 kHz

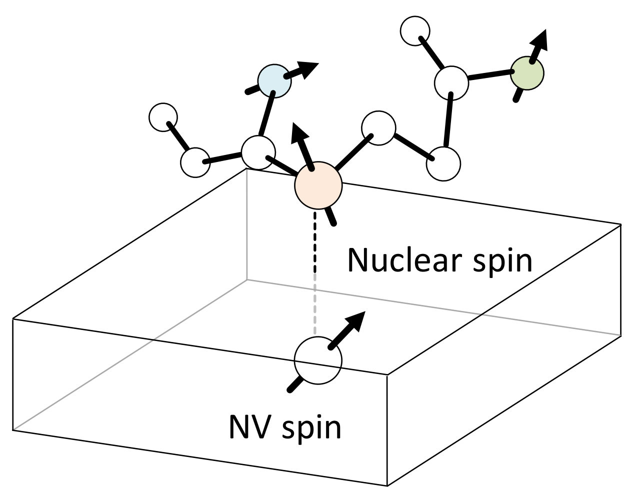
DFT: New J. Phys. **20**, 023022 (2018) Nizovtsev *et al.*

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

Toward single-molecular imaging

- **Information of the positions of the individual nuclei**
  → Accurate measurement of *e*–*n* int. const's $(A_{\parallel}, A_{\perp}) \approx (r, \theta)$
  → Lack of information on the azimuthal angle ϕ

- **Spectral resolution**
  → Easy to resolve isotopes
  → Need to measure *J*-couplings & chemical shifts (ppm!)
  → Limited by sensor/memory lifetimes $(T_{2e/n}, T_{1e/n})$



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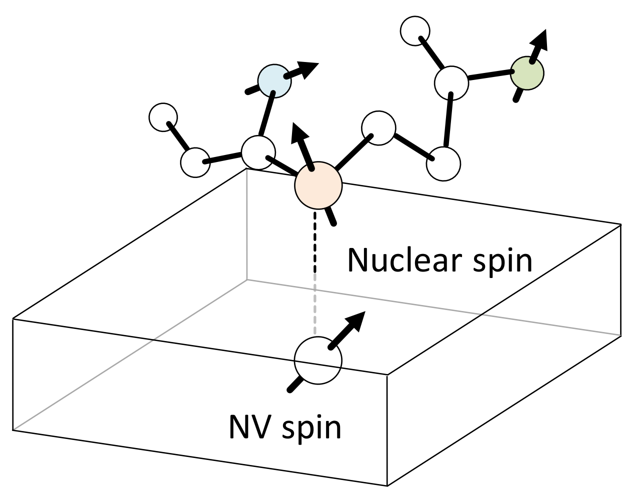
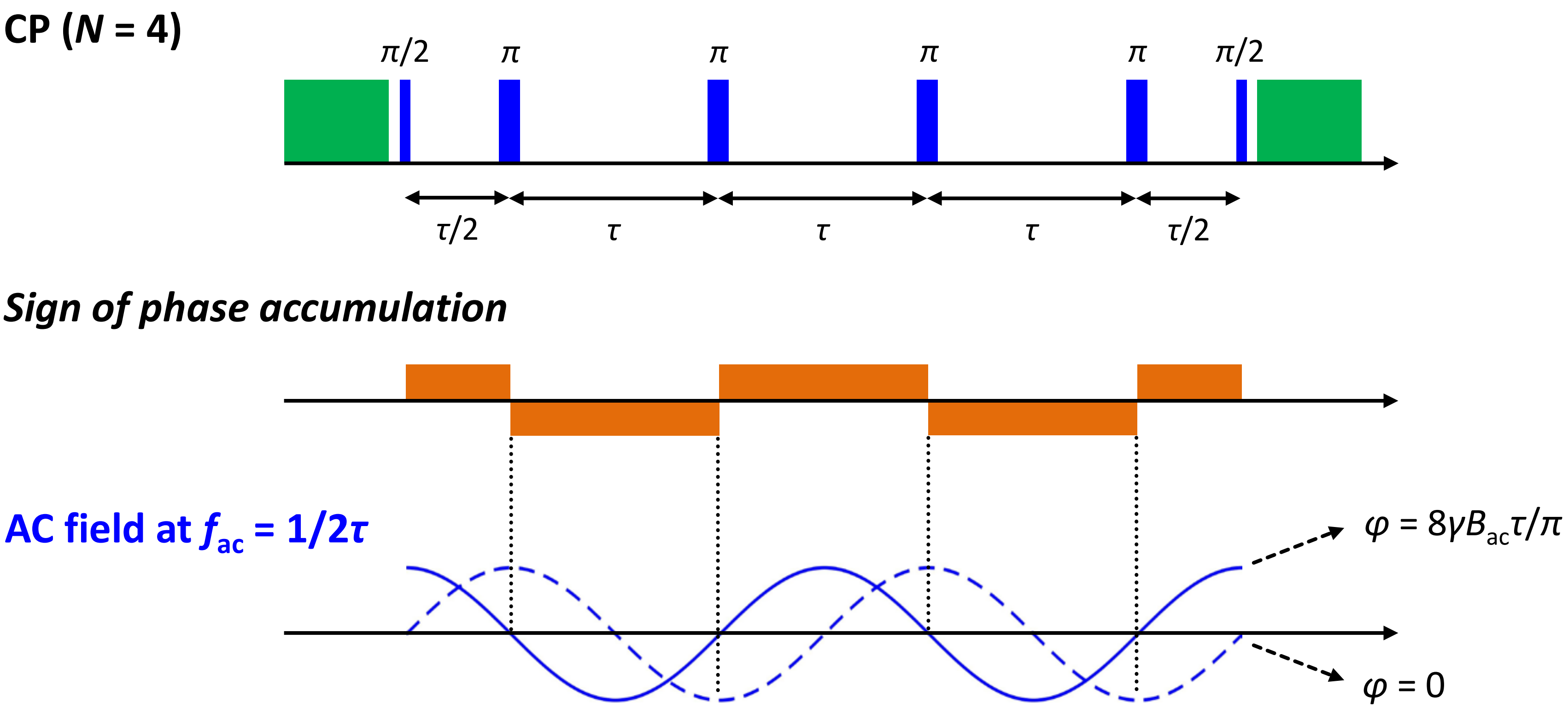
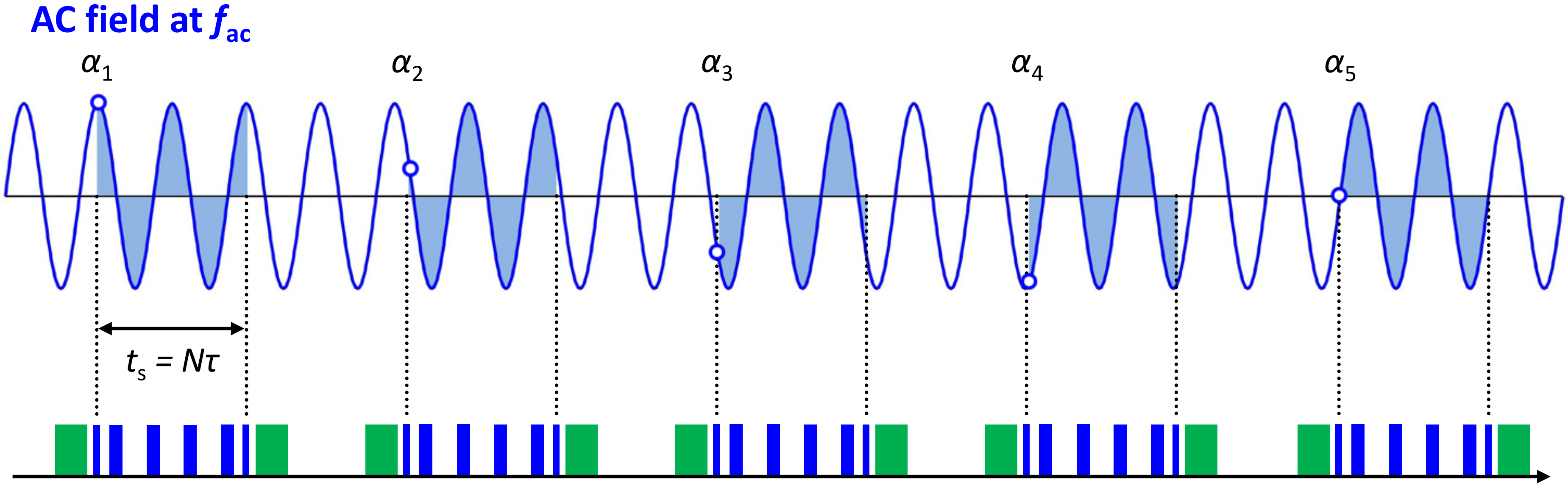


AC magnetometry



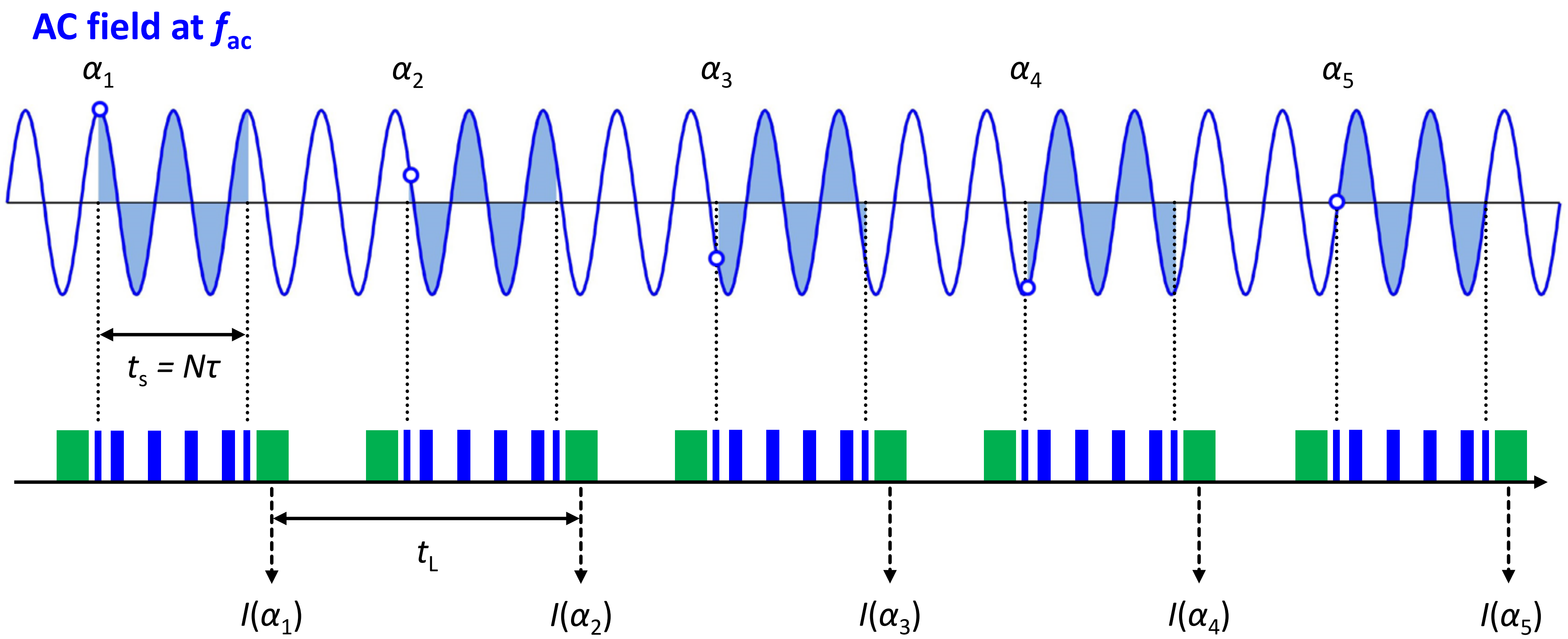
- φ depends on the **initial phase α of the AC field** ($\varphi \propto \cos\alpha$)

AC magnetometry



- φ depends on the **initial phase $\boldsymbol{\alpha}$ of the AC field** ($\varphi \propto \cos \alpha$)
- Average over **random $\boldsymbol{\alpha}$**

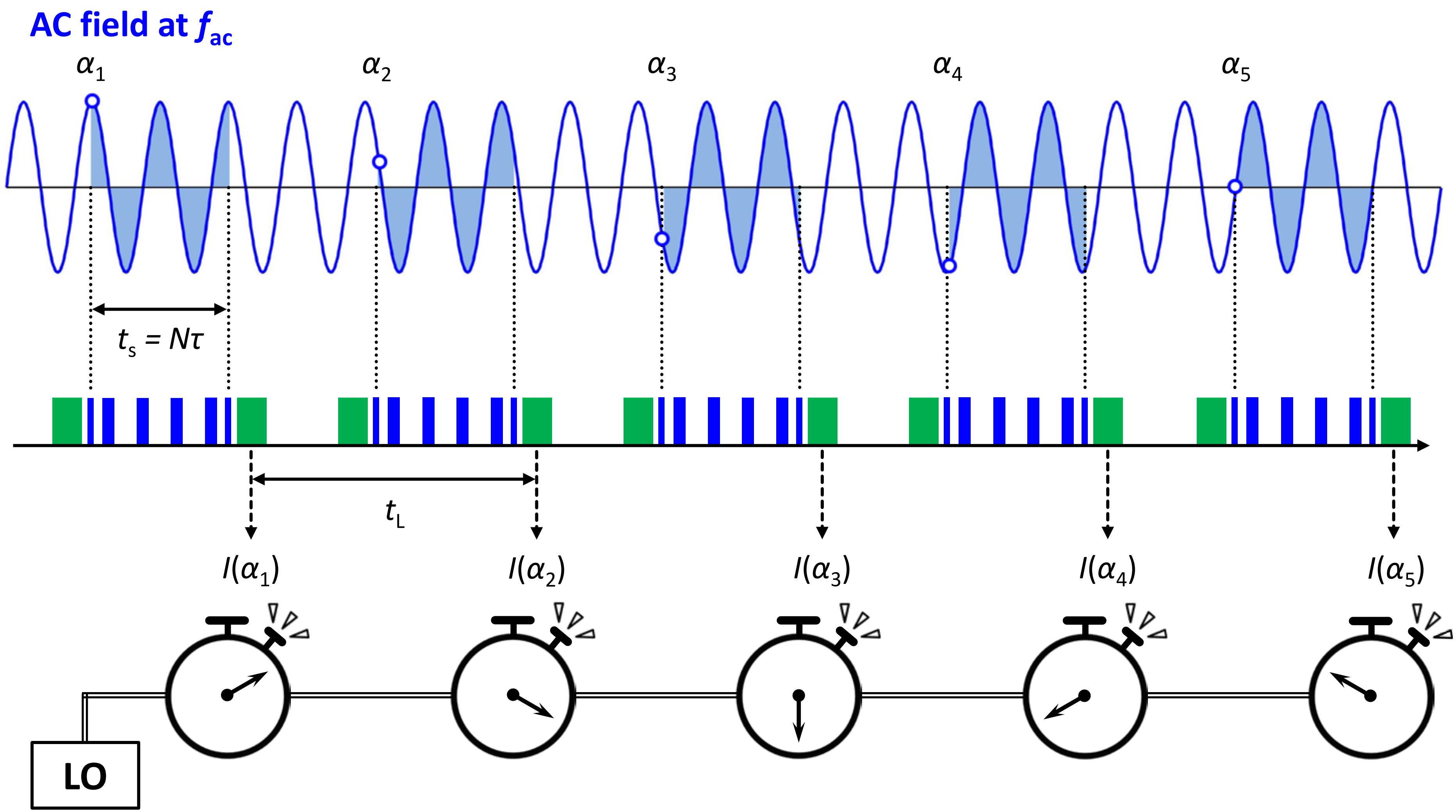
Ultrahigh resolution sensing



- φ depends on the **initial phase $\boldsymbol{\alpha}$ of the AC field** ($\varphi \propto \cos\alpha$)
- Average over **random $\boldsymbol{\alpha}$**
- **If the data acq. is periodic,** adjacent α's are related by $\alpha_{k+1} = 2\pi f_{ac} t_L + \alpha_k$

Science **356**, 832 (2017) Schmitt *et al.*; Science **356**, 837 (2017) Boss *et al.*; Nature **555**, 351 (2018) Glenn *et al.*

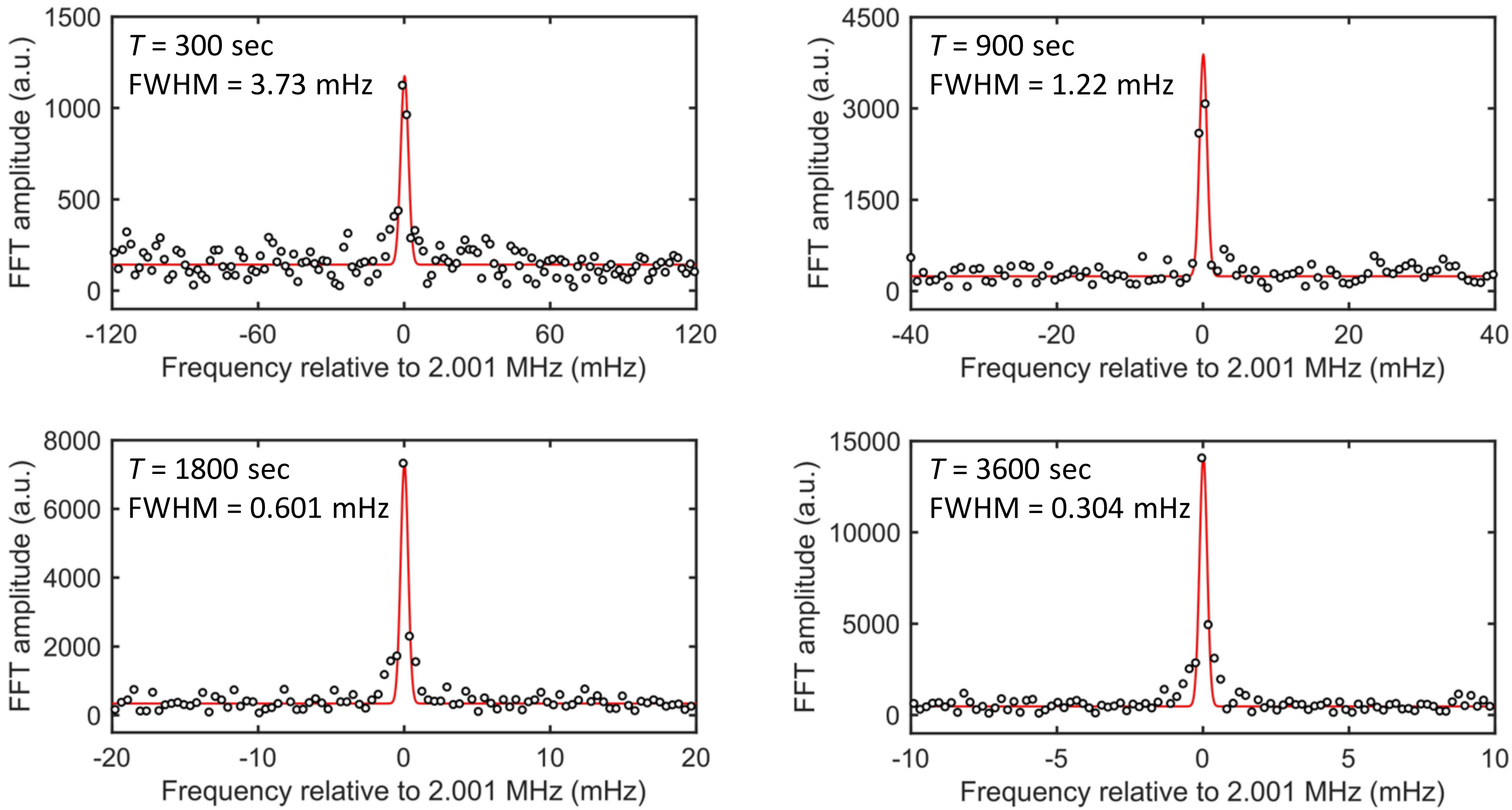
Ultrahigh resolution sensing



Science **356**, 832 (2017) Schmitt *et al.*; Science **356**, 837 (2017) Boss *et al.*; Nature **555**, 351 (2018) Glenn *et al.*

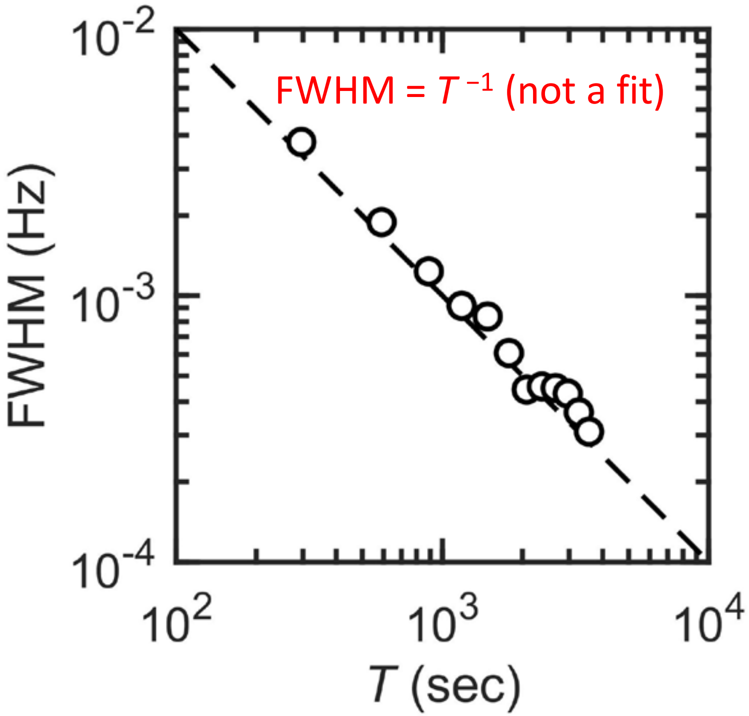
Ultrahigh resolution sensing

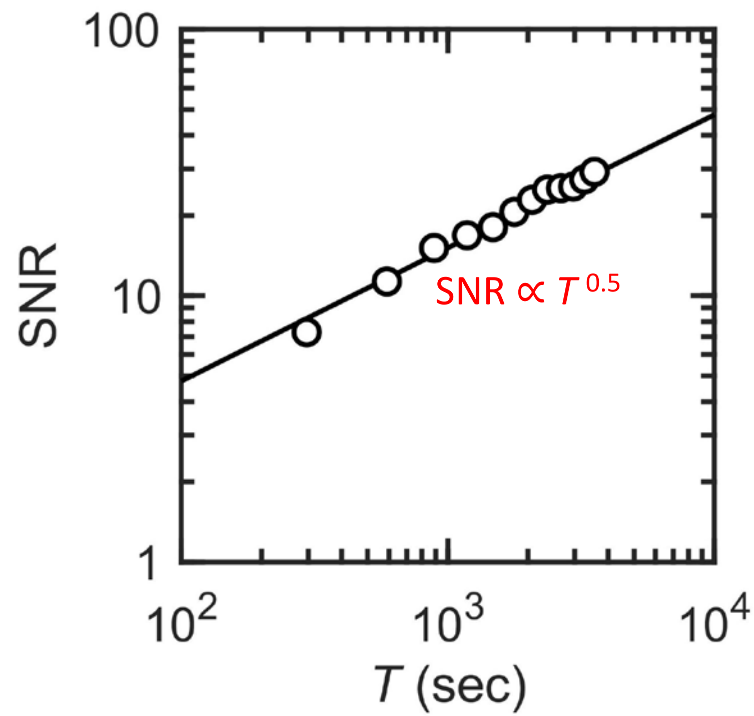
B_{ac} = 96.5 nT & f_{ac} = 2.001 MHz applied from a coil, detected by a single NV center



J. Appl. Phys. **123**, 161101 (2018) Abe & Sasaki

Ultrahigh resolution sensing



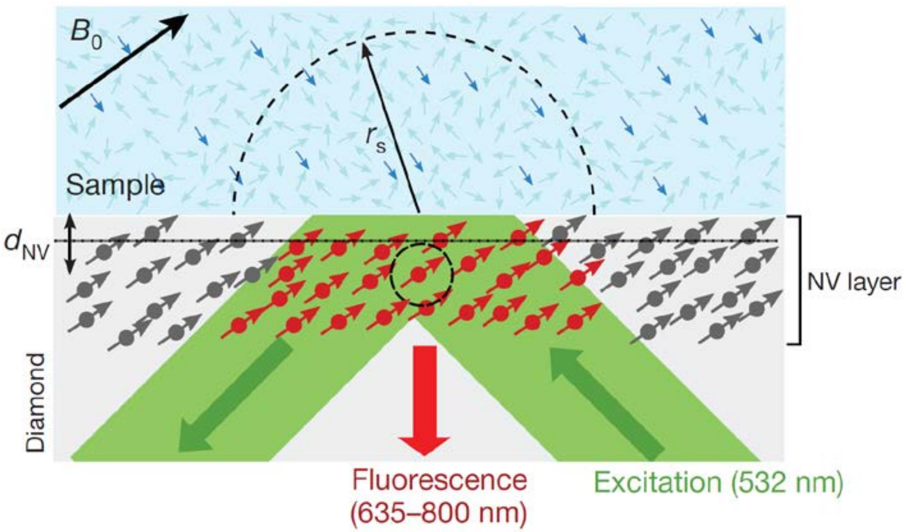


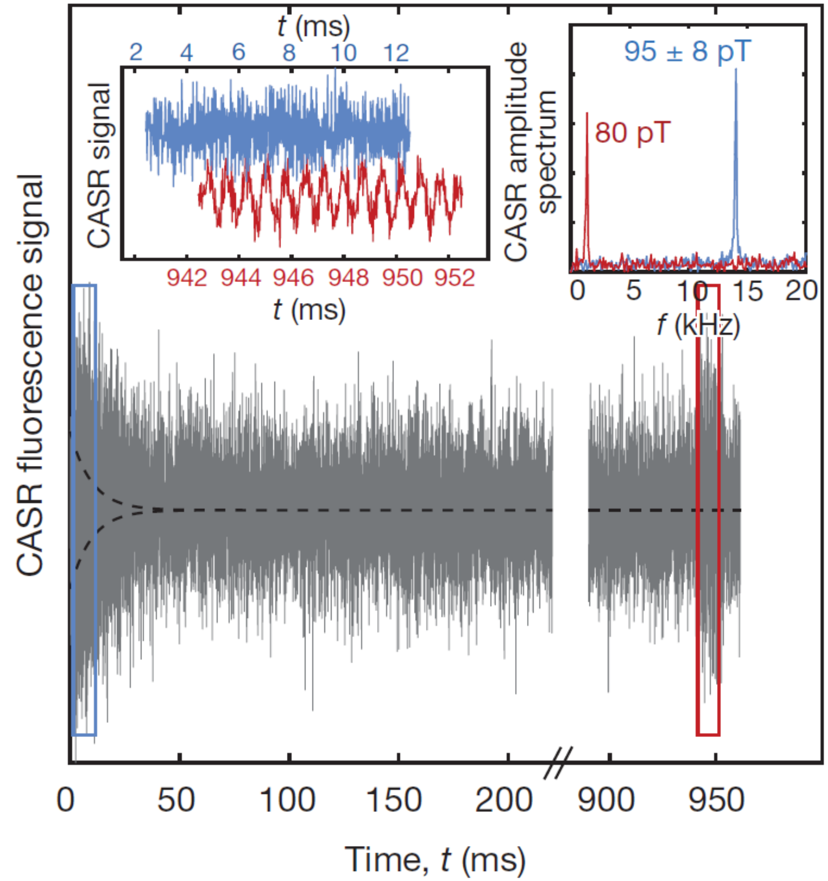
- Spectral resolution not limited by sensor/memory lifetimes ($T_{2e/n}$, $T_{1e/n}$)
- Only limited by the stability of LO (essentially infinite)
- Resolution = T^{-1} & SNR ∝ $T^{0.5}$ → Precision ∝ $T^{-1.5}$

J. Appl. Phys. **123**, 161101 (2018) Abe & Sasaki

NMR spectroscopy

Data from Harvard: Nature **555**, 351 (2018) Glenn *et al.*





- [NV] ≈ 3 x 10^{17} cm^{-3}
- # of NV ≈ 5 x 10^9
- V_{detect} ≈ 25 pL
- # of protons ≈ 2.5 x 10^{15}
- RF pulse → FID

See also: Science **357**, 67 (2017) Aslam *et al.* (Wrachtrup, Stuttgart)
[B_0 = 3 T, f_e = 87 GHz, T_{1n} = 260 s]

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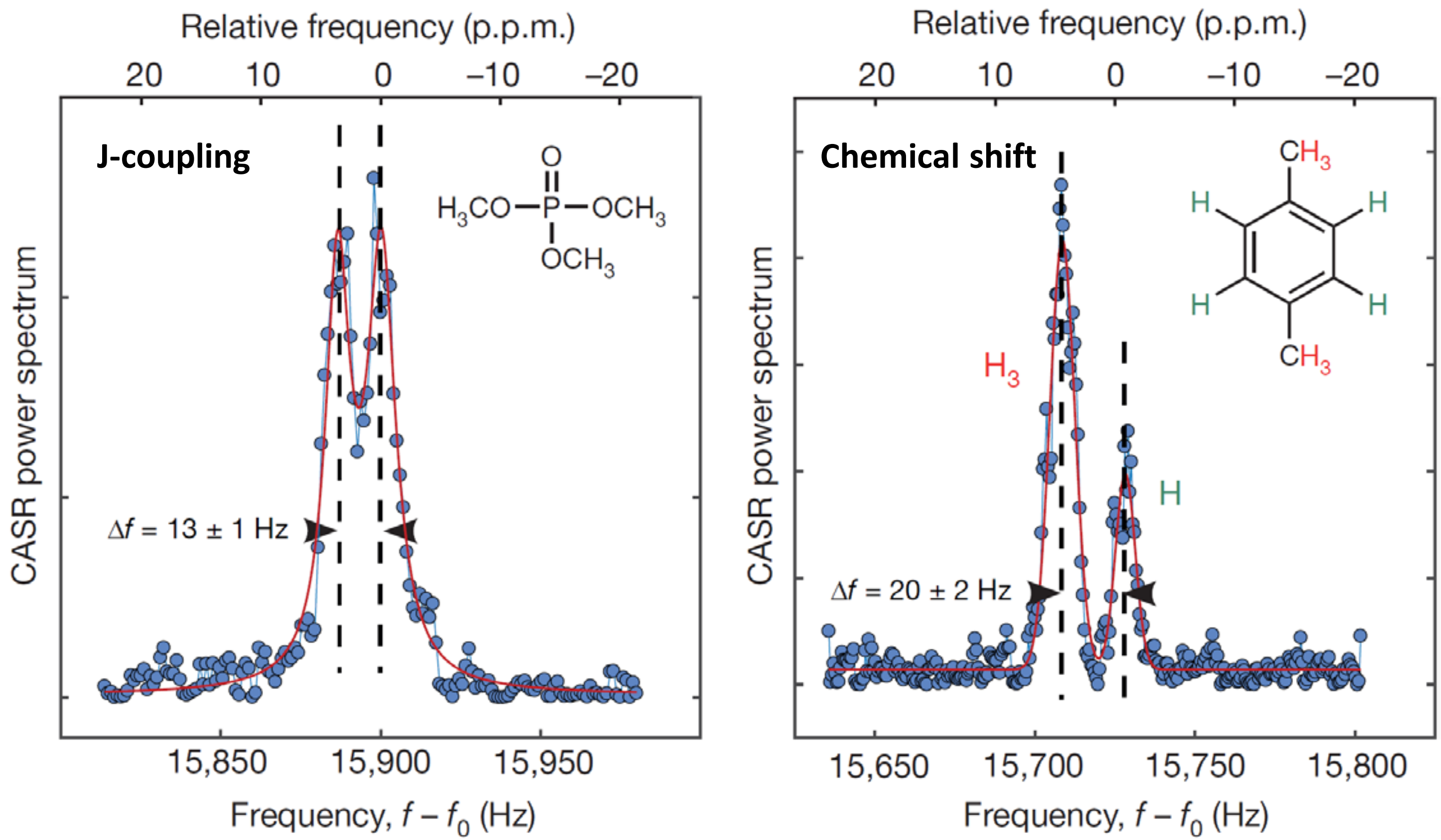

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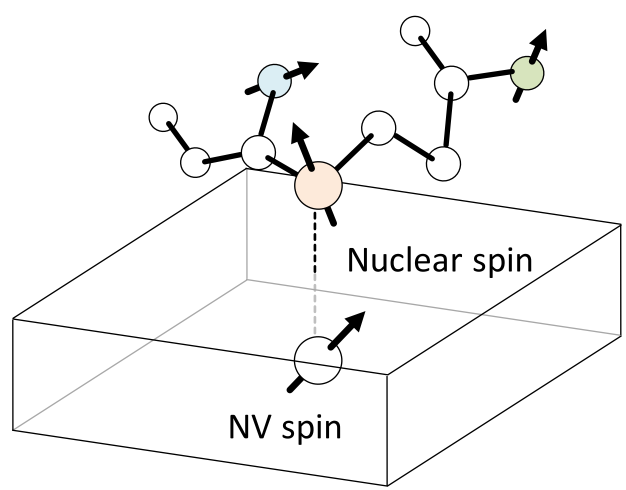
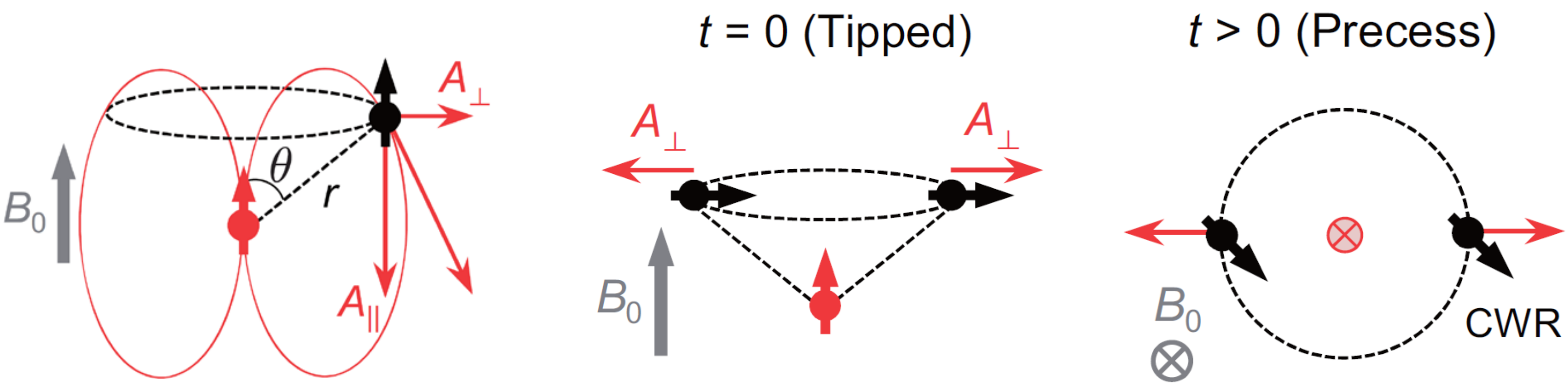


How to determine ϕ?



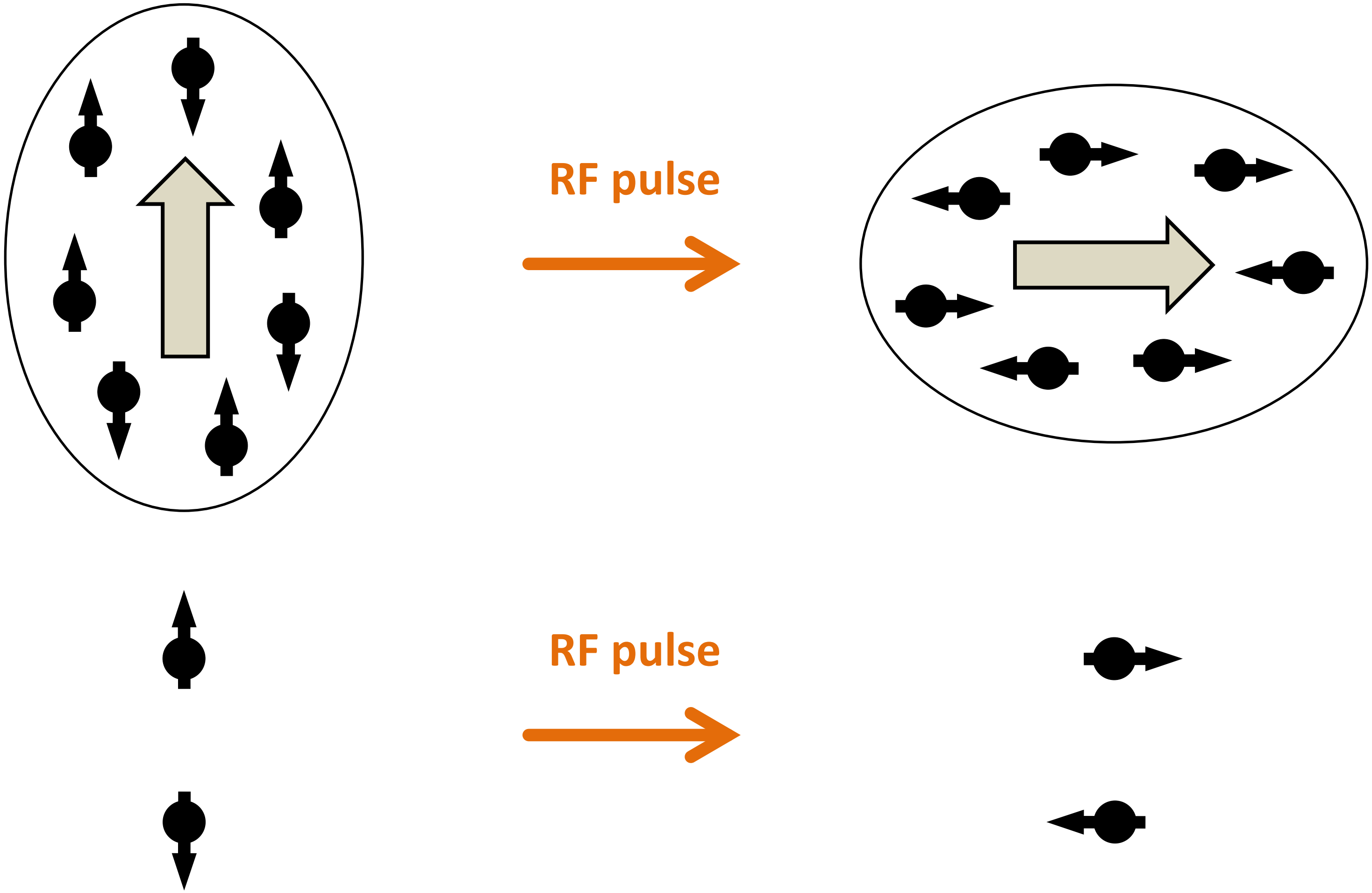
Transition probability of the NV spin after the detection of a single nuclear spin

$$P_Y = \frac{1}{2} - \frac{1}{2}\cos(\phi - \phi_n)\sin N\phi_{cp}$$

Azimuthal angle of the nuclear Bloch vector: $2\pi f_p t + \phi_n(0)$

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

Ensemble vs. single

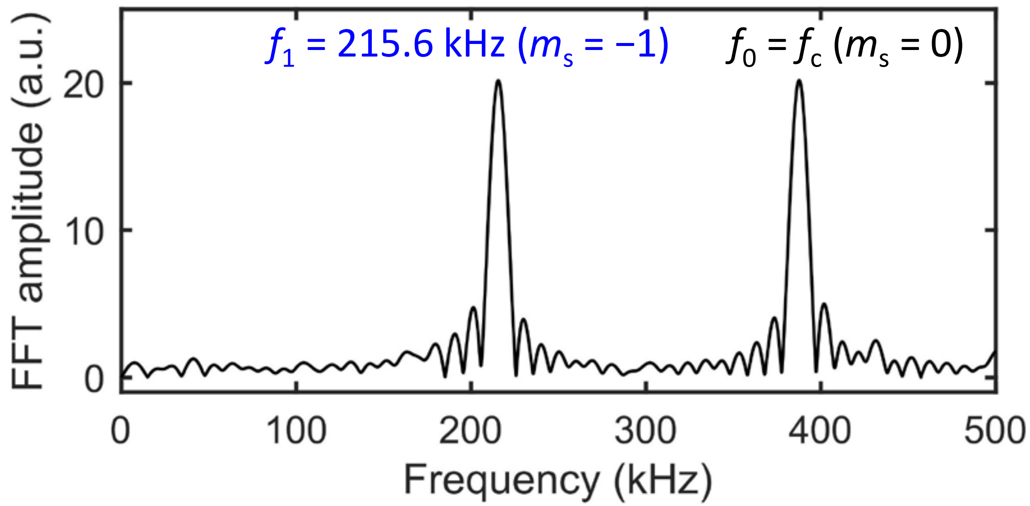


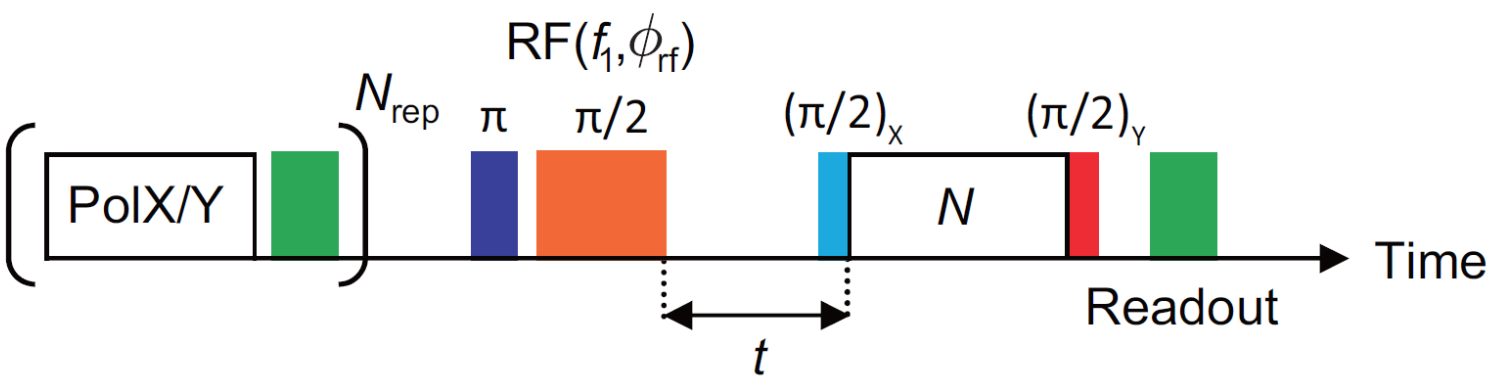
The initial state matters

→ Dynamic nuclear polarization (DNP)

Determination of ϕ of a ^{13}C n-spin

1. **DNP (PulsePol)**
2. **RF pulse@$m_s = -1$**
3. **Wait t (n-spin precesses)**
4. **AC sensing**



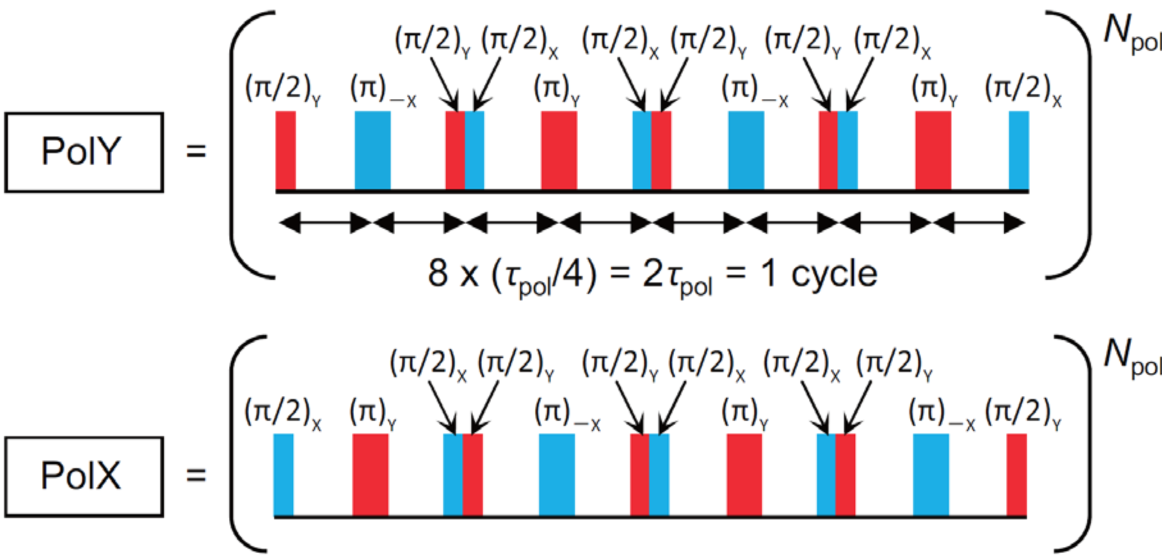


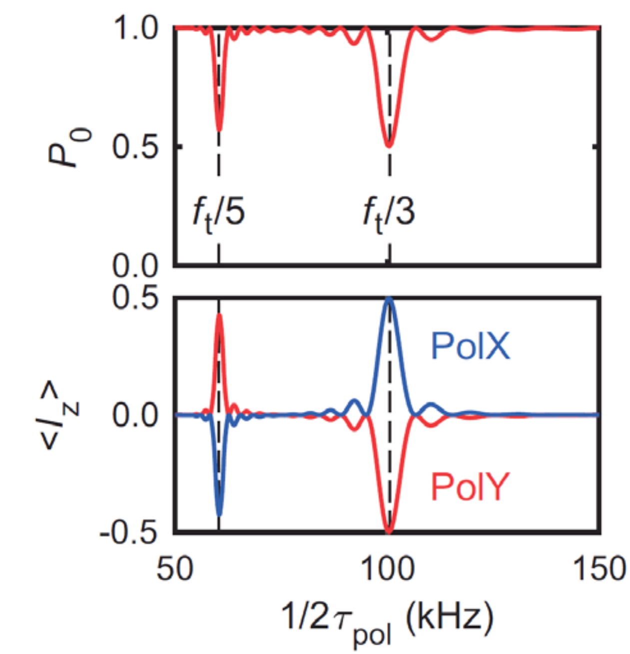
Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

PulsePol

Hamiltonian engineering

- DNP condition: $2\tau_{pol} = k/f_n$ (k: odd, f_n: nuclear Larmor frequency)
- Average Hamiltonian $\propto S_+I_- + S_-I_+, \propto S_+I_+ + S_-I_-$

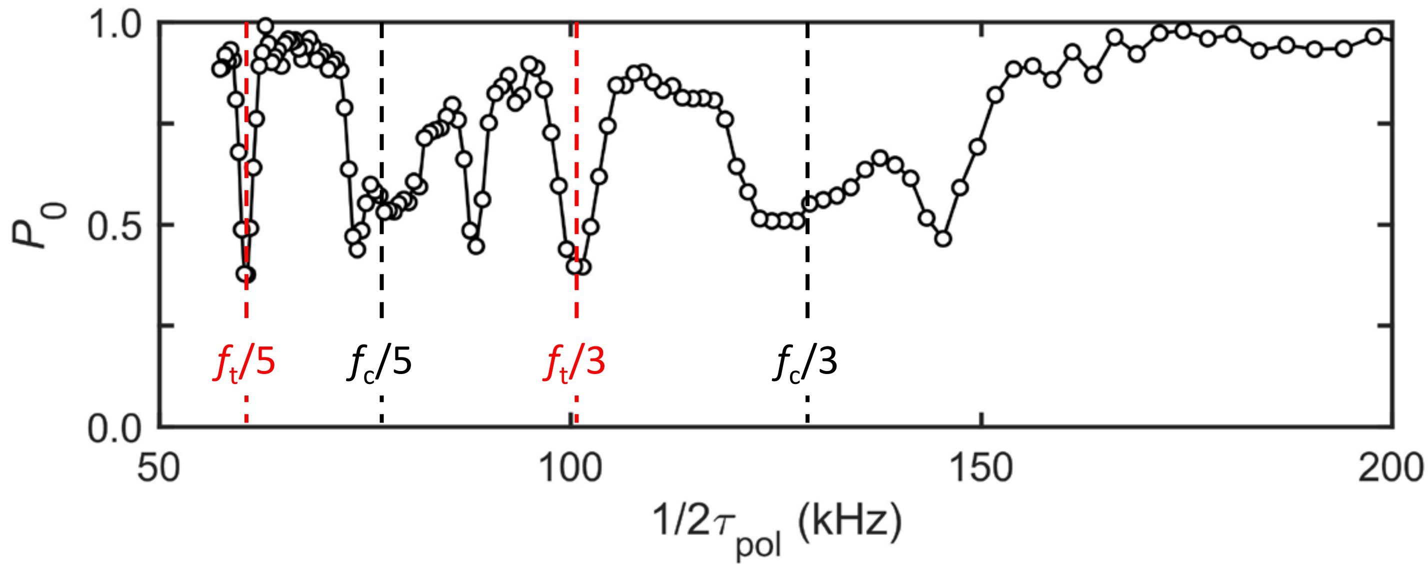


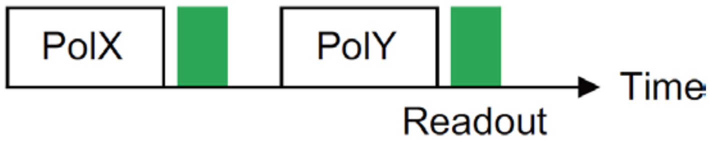


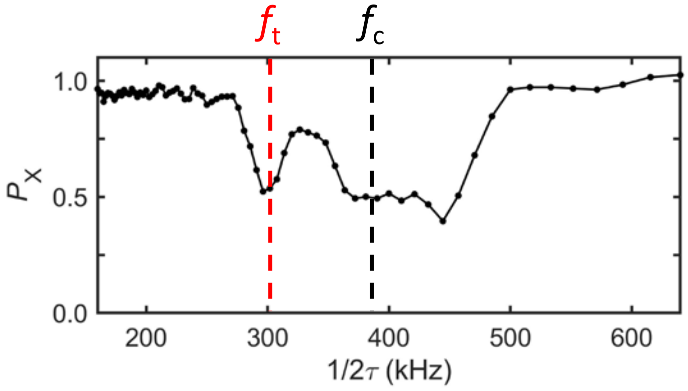
[PulsePol] Sci. Adv. **4**, eaat8978 (2018) Schwartz *et al.*

Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

PulsePol

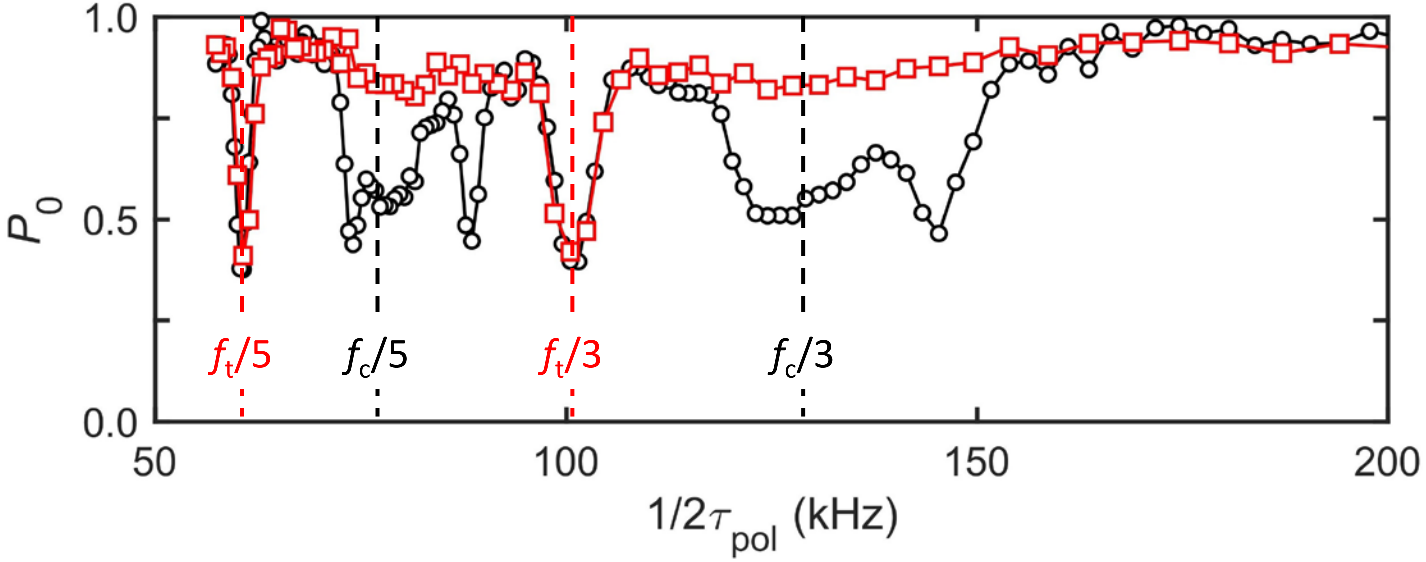


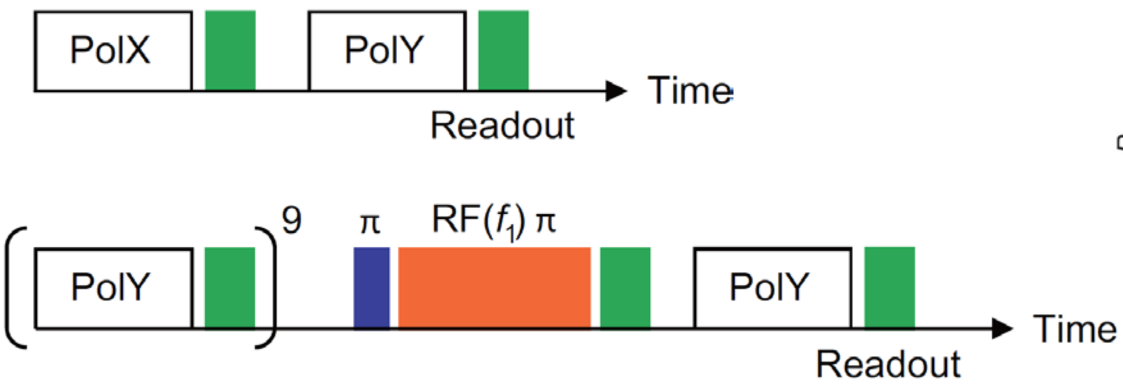


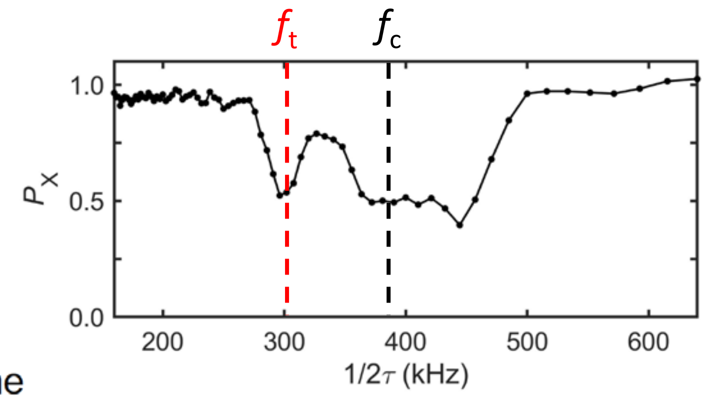


Phys. Rev. B **98**, 121405 (2018) Sasaki *et al.*

PulsePol







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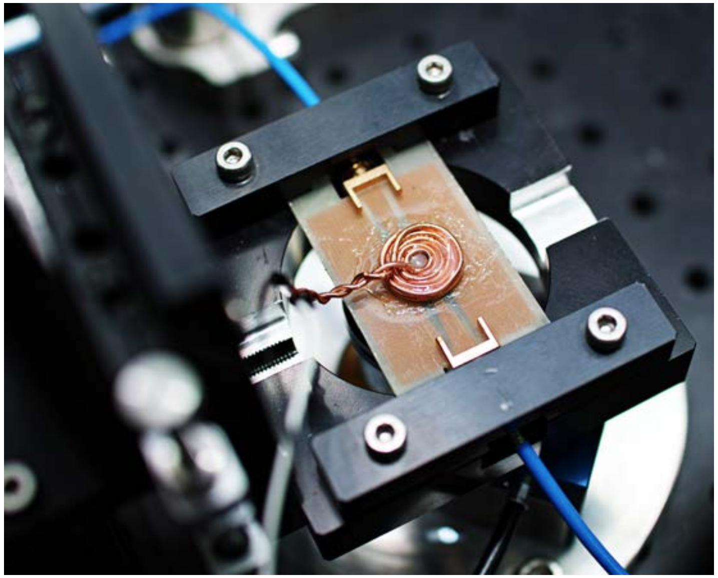
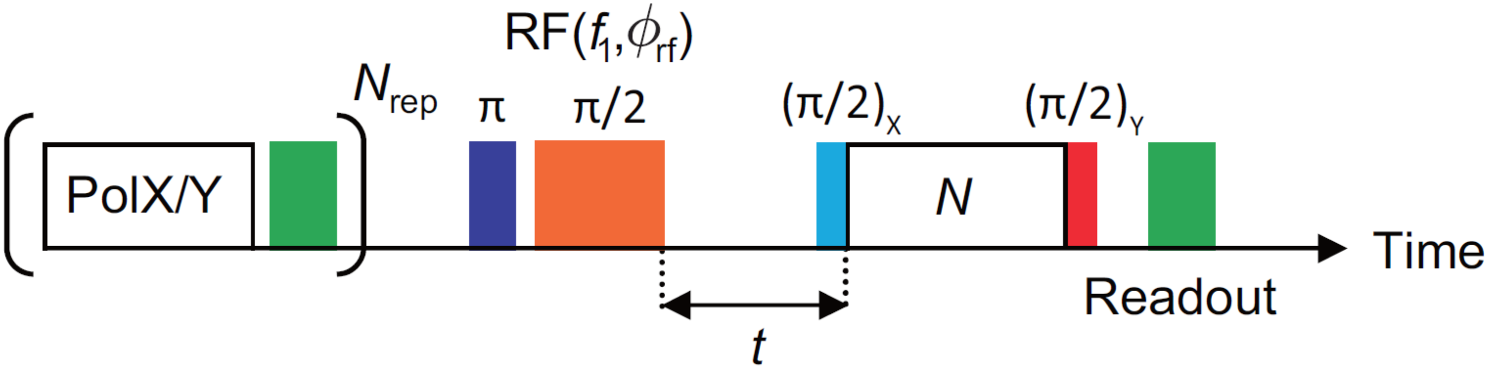
Determination of ϕ of a ^{13}C n-spin

1. **DNP (PulsePol)**
2. **RF pulse@m_s = −1**
3. **Wait *t* (*n*-spin precesses)**
4. **AC sensing**





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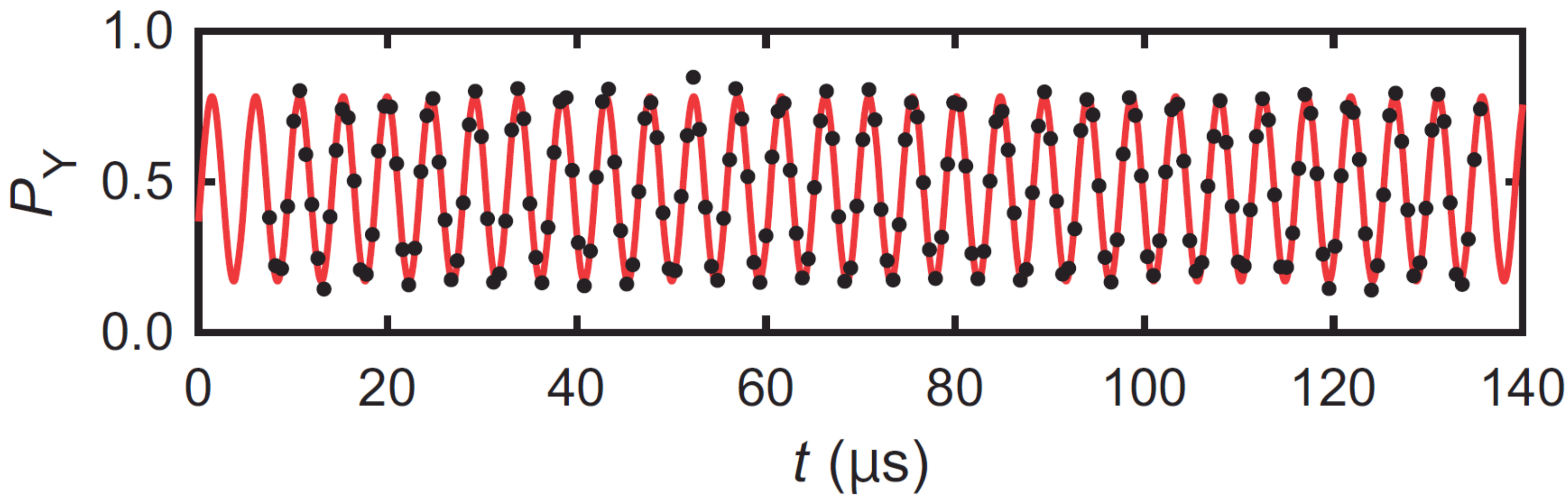
Determination of ϕ of a ^{13}C *n*-spin



- ✓ $t \rightarrow$ 1 ms (undersampling)
- ✓ f_p = 215.79 kHz ≈ f_1 = 215.6 kHz
- ✓ $\phi - \phi_n(0) = 334.0^o$
- ✓ $\phi_n(0) = 89.2^o$ (Real-space *n*-spin trajectory)

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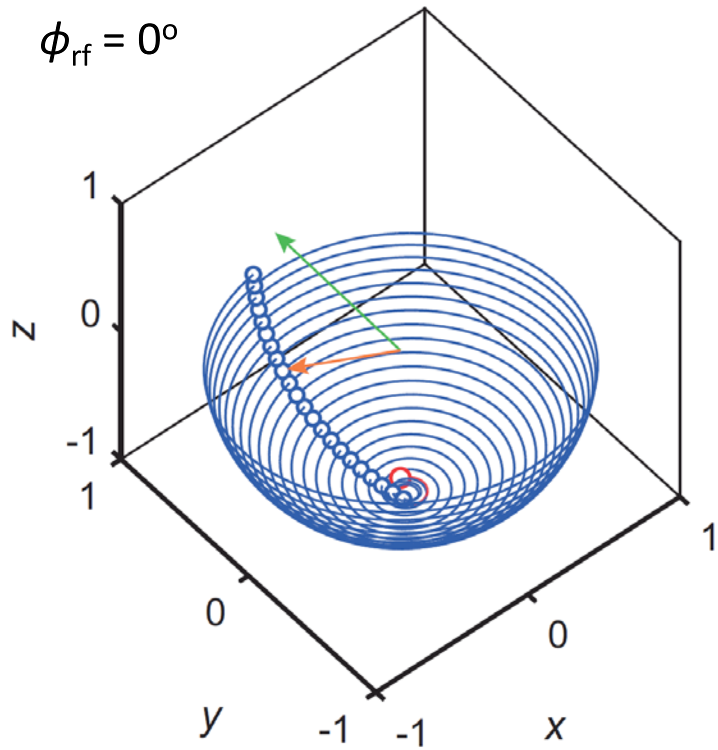
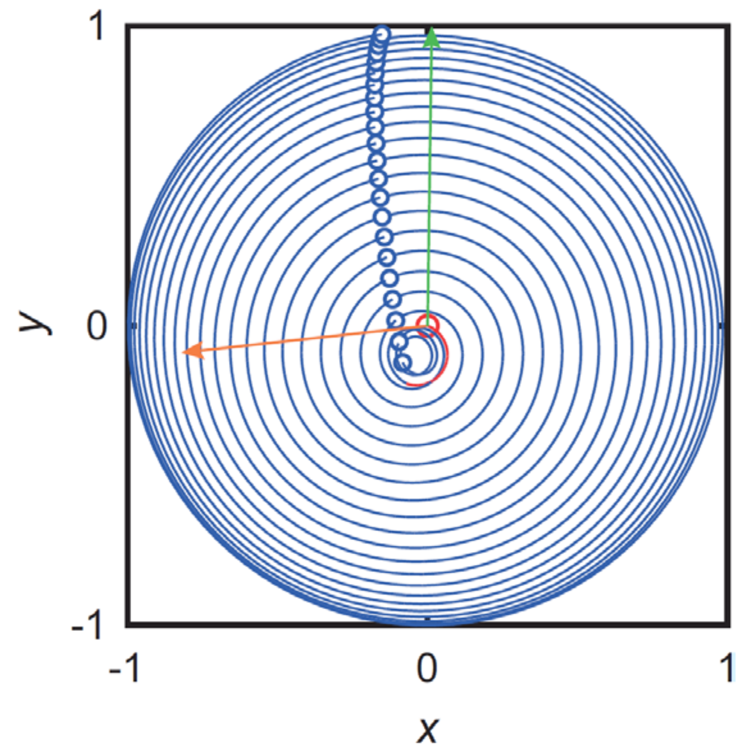
$\phi_n(0)$: Real-space *n*-spin trajectory





Direction of the RF field

Bloch vector@t = 0 (Considering the rotation axis of the n-spin & detuning)

Full simulation based on the Bloch equation (Tilt of q-axis, pulse delay...)

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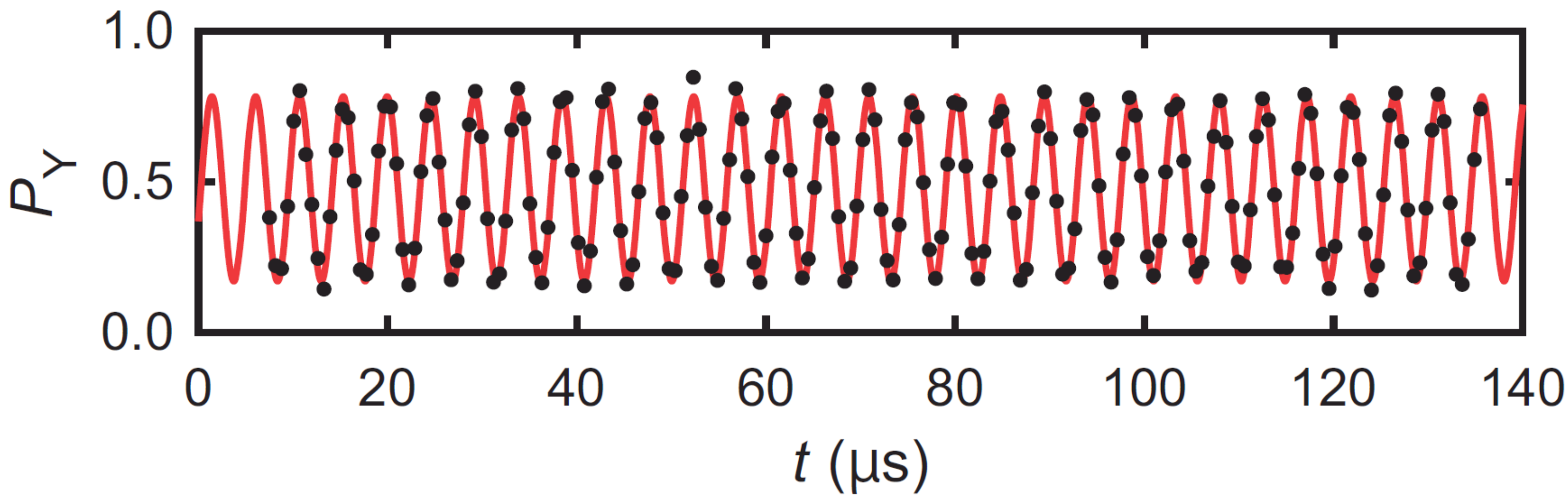
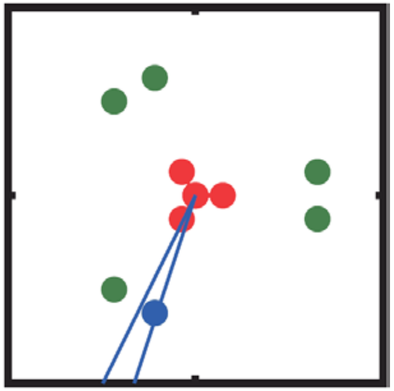
Determination of ϕ of a ^{13}C *n*-spin



- ✓ $t \rightarrow$ 1 ms (undersampling)
- ✓ f_p = 215.79 kHz ≈ f_1 = 215.6 kHz
- ✓ $\phi - \phi_n(0) = 334.0^o$
- ✓ $\phi_n(0) = 89.2^o$ (Real-space *n*-spin trajectory)

→ $\phi = 247.8 \pm 4.1^o$



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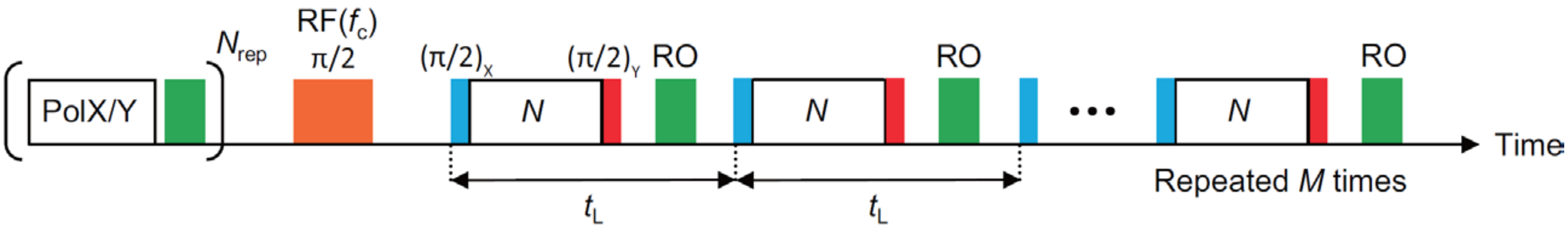
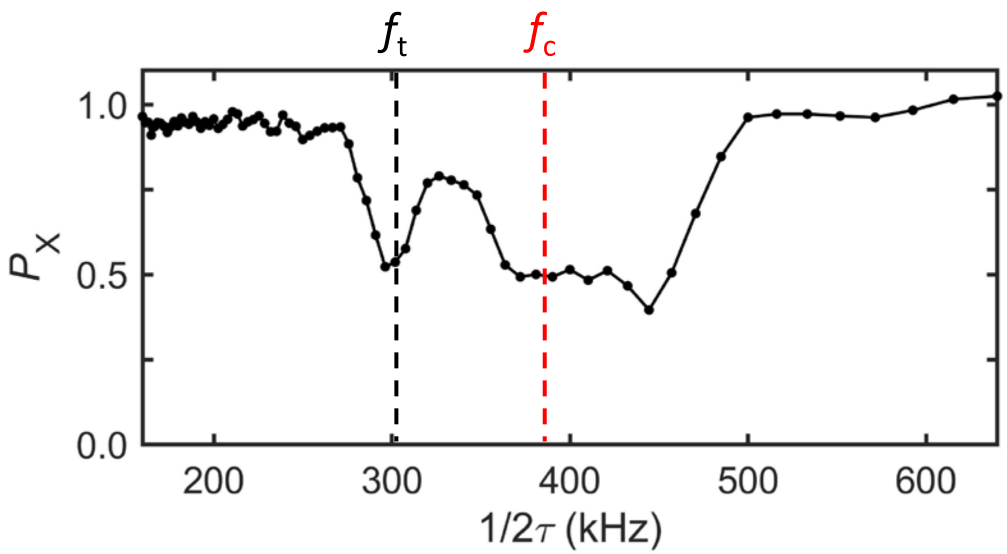
Observation of weakly coupled ^{13}C *n*-spins

Ultrahigh resolution sensing (*N* = 2)





- f_c = 387.5 kHz
- RF pulse@m_s = 0
- N = 2 to minimize **back actions** from NV

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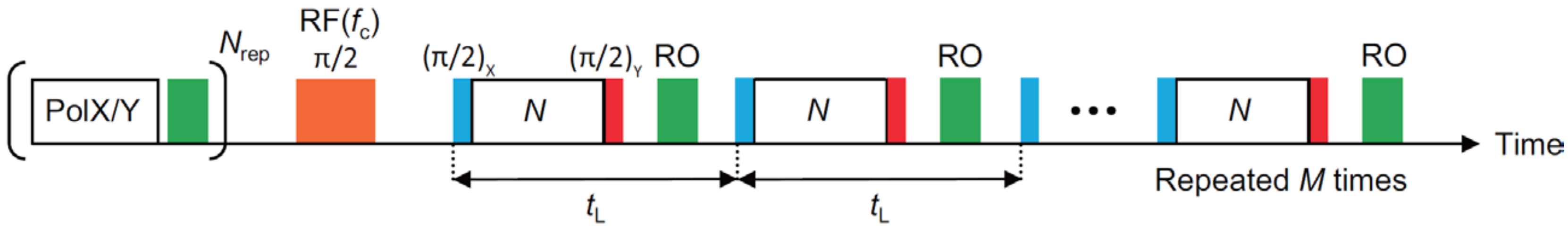
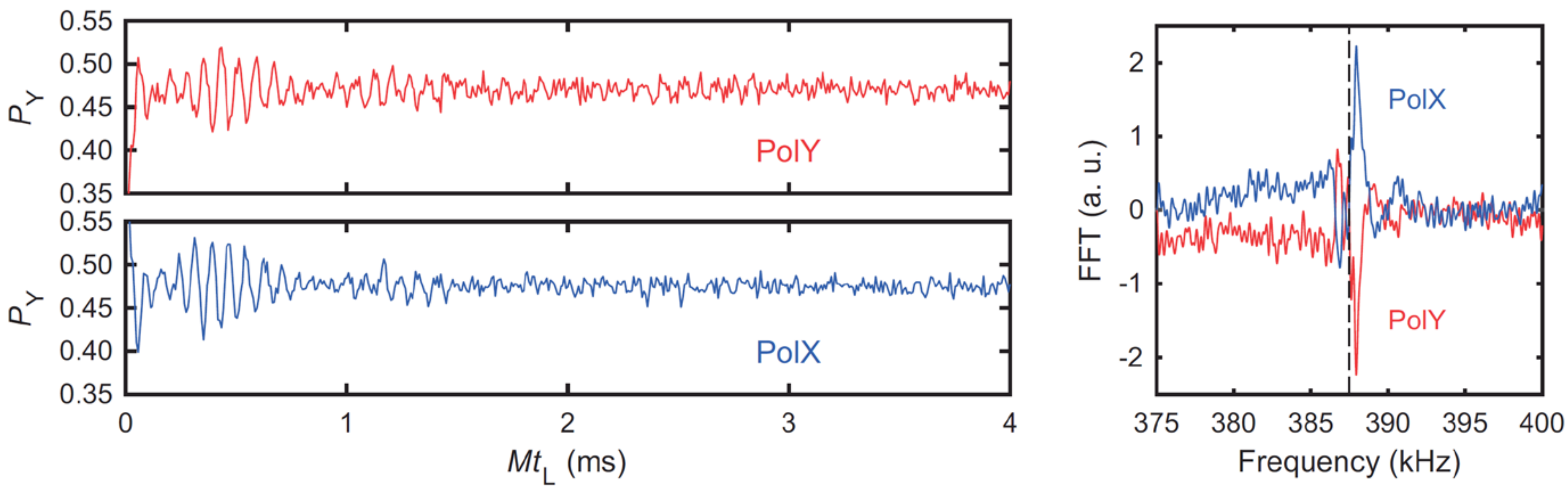
Observation of weakly coupled ^{13}C n-spins

Ultrahigh resolution sensing (N = 2)





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Summary

- **Tools for single-molecule imaging/structural analysis are being developed**

  → Determination of the position of a single *n*-spin[1,2]
  → Ultrahigh resolution sensing on single *n*-spins[1,3,4]

- **Other issues:** Create high-quality "shallow" NVs, accurately position single molecules/proteins near the sensor etc

[1] Phys. Rev. B **98**, 121405 (2018) Sasaki, Itoh & Abe (arXiv.1806.00177)
[2] Phys. Rev. Lett. **121**, 170801 (2018) Zopes *et al.* (arXiv.1807.04559)
[3] Nature Commun. **10**, 594 (2019) Pfender *et al.* (arXiv.1806.02181)
[4] Nature **571**, 230 (2019) Cujia *et al.* (arXiv.1806.08243)

→ Suppressing back actions by weak measurement