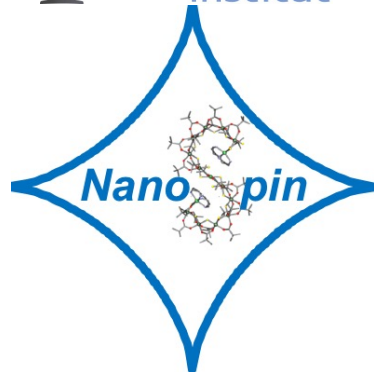


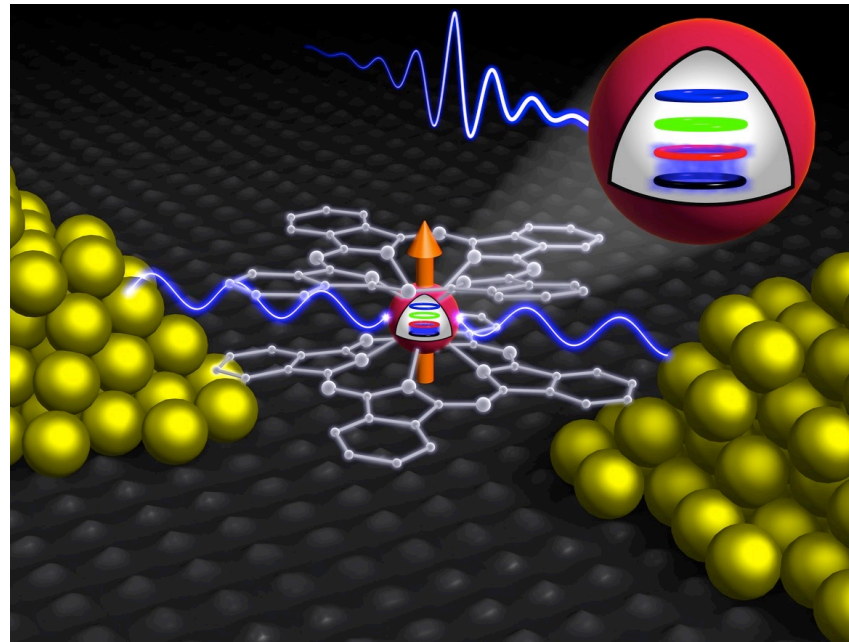
Nano Quantum Magnetism

W. Wernsdorfer

Karlsruhe Institute of Technology (PHI, INT)
Institut Néel, CNRS, Grenoble



European
Research
Council



Physikalisches
Institut



Lecture 10



Alexander von Humboldt
Stiftung / Foundation

Outline

Di Vincenzo 1996



Isolation



Read-out



Initialization



Manipulation



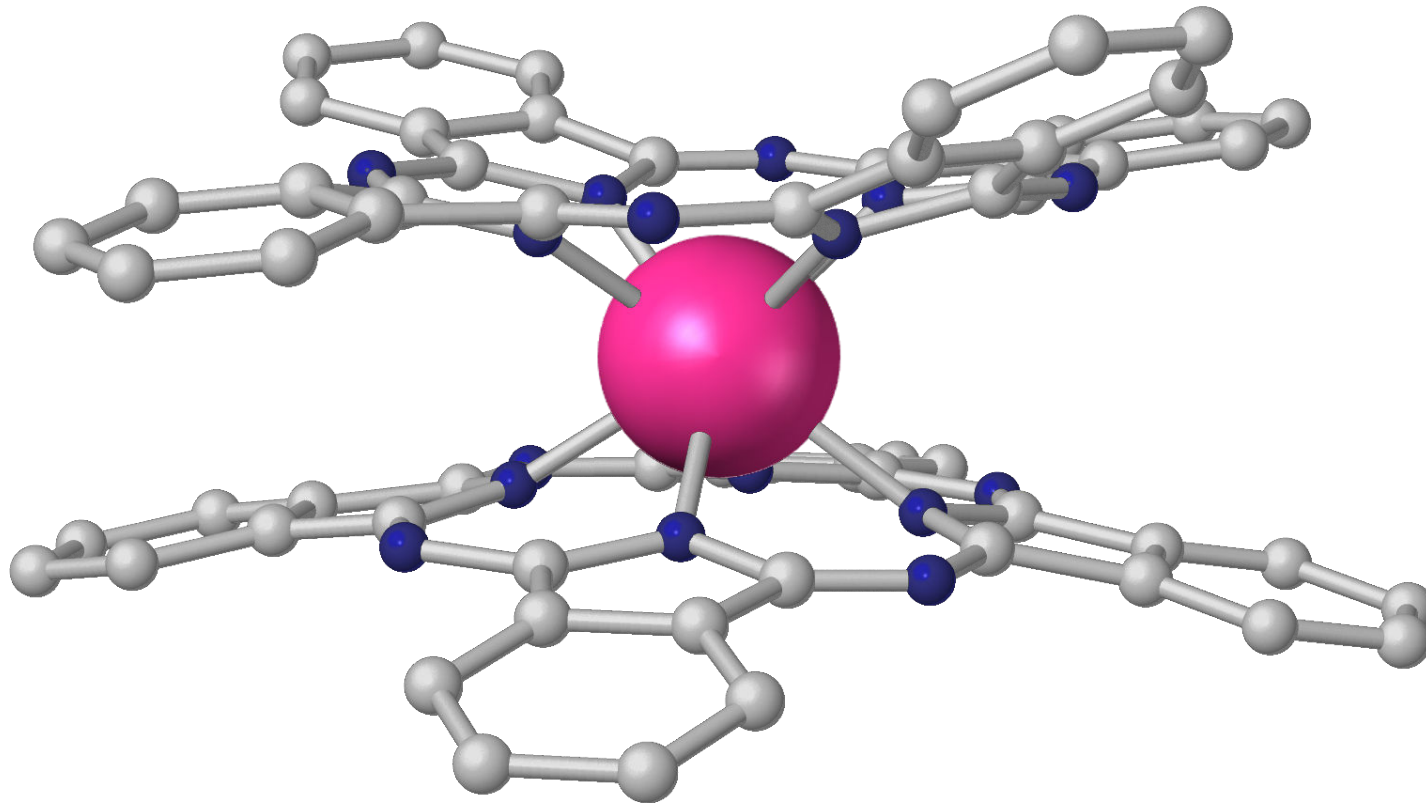
Storage



Quantum gates, Grover's algorithm

Nuclear spin-qubit SMM

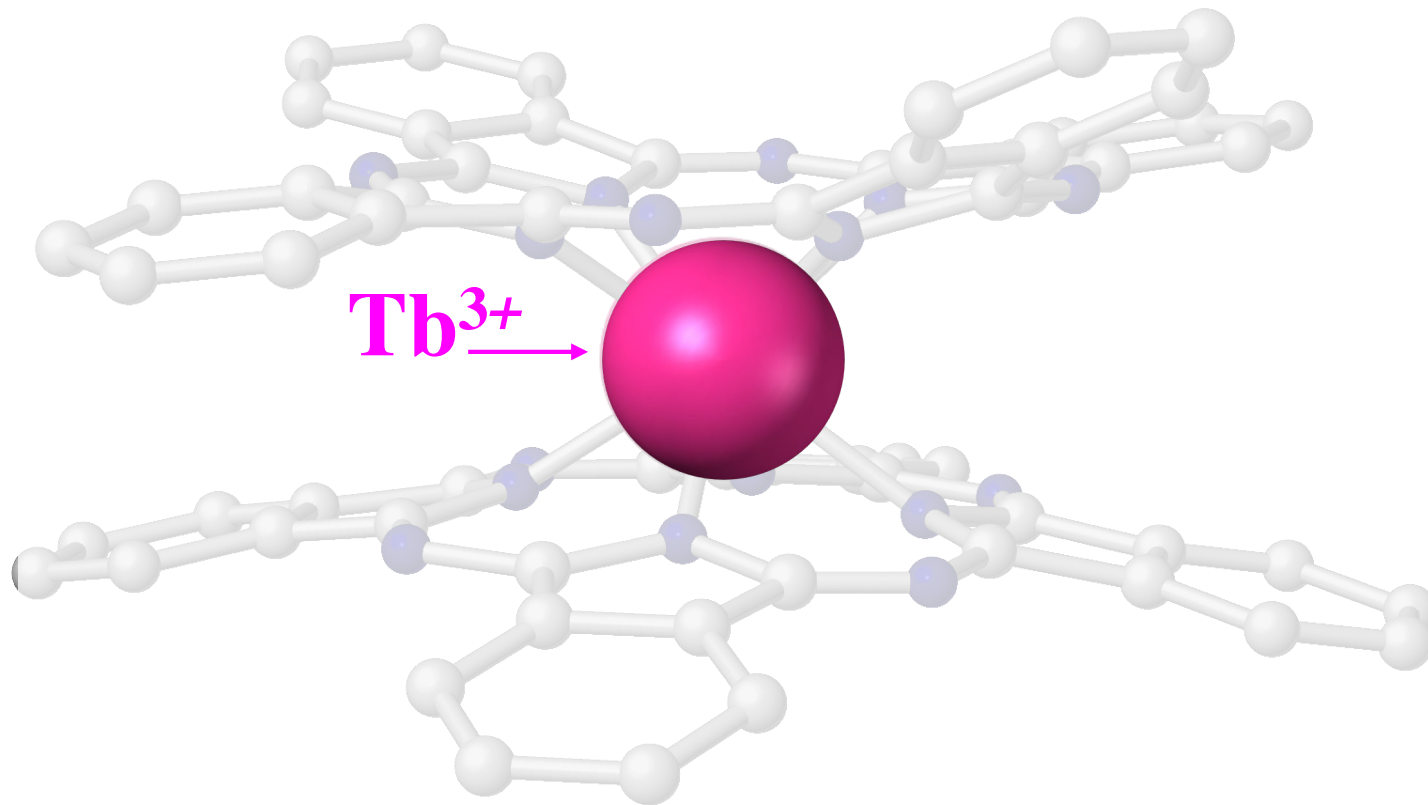
- terbium double-decker (TbPc_2) – a simple example



Jmol

Nuclear spin-qubit SMM

- terbium double-decker (TbPc_2)



Jmol

Nuclear spin-qubit SMM

- terbium double-decker (TbPc_2)

electronic spin

- $J = 6$

- $\mu \approx 10 \mu_B$

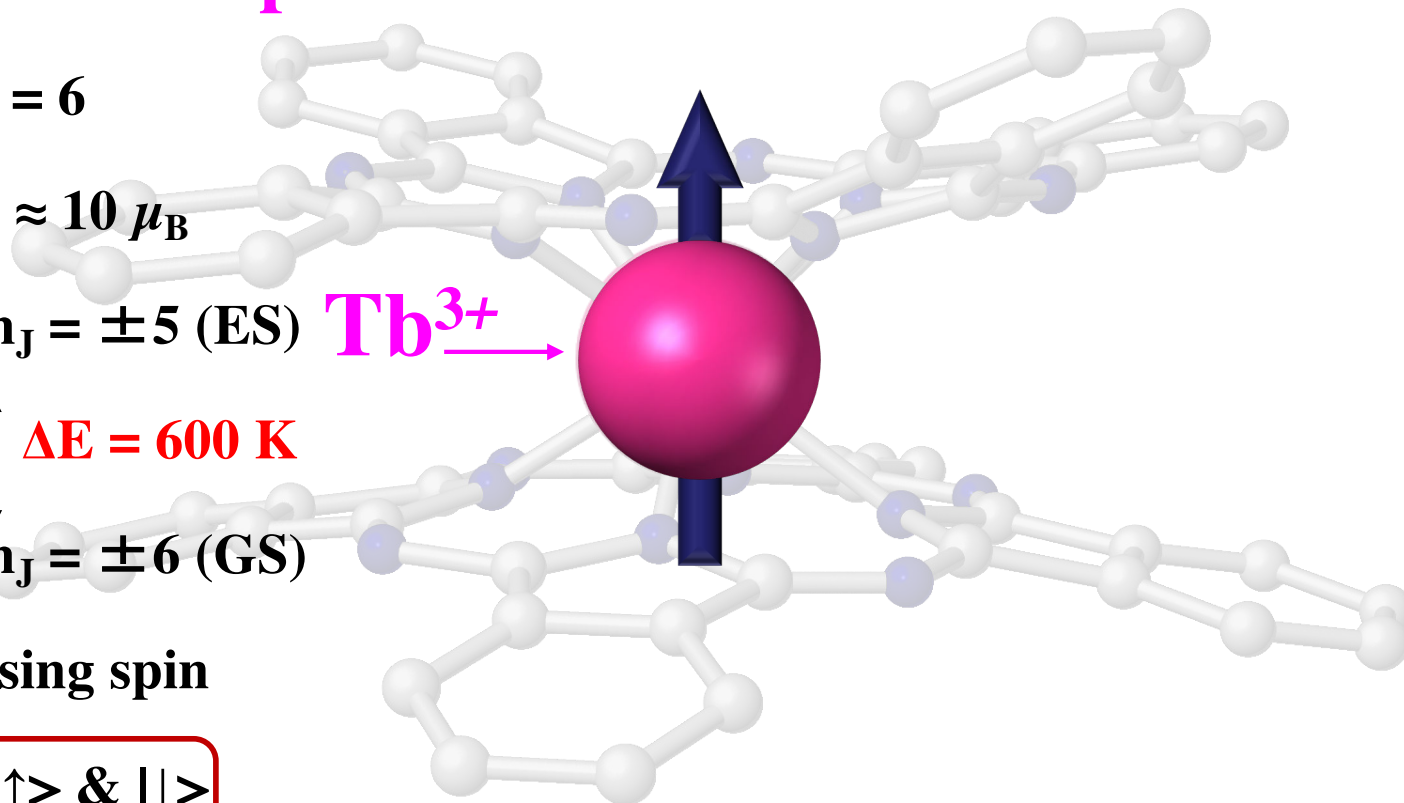
- $m_J = \pm 5$ (ES) Tb^{3+}

- $\Delta E = 600 \text{ K}$

- $m_J = \pm 6$ (GS)

→ Ising spin

$|\uparrow\rangle$ & $|\downarrow\rangle$



Jmol

Nuclear spin-qubit SMM

- terbium double-decker (TbPc_2)

electronic spin

- $J = 6$
- $\mu \approx 10 \mu_B$
- $m_J = \pm 5$ (ES) Tb^{3+} \rightarrow
- \updownarrow $\Delta E = 600 \text{ K}$
- $m_J = \pm 6$ (GS)

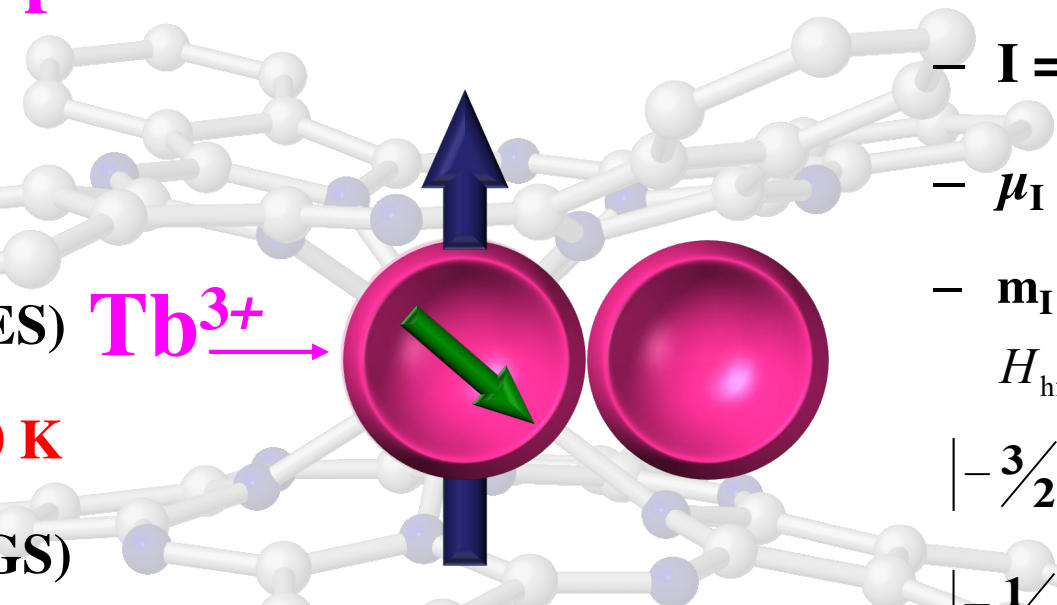
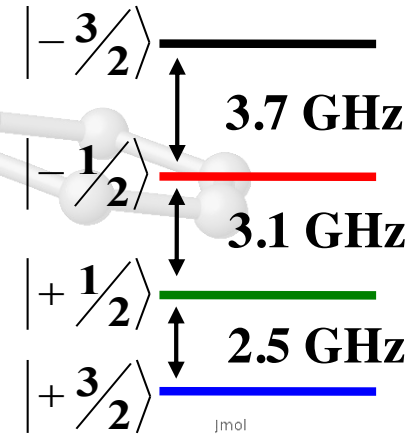
\rightarrow Ising spin

$|\uparrow\rangle \ \& \ |\downarrow\rangle$

environmental
isolation!

nuclear spin

- $I = 3/2$
 - $\mu_I \approx 1/1000 \mu_B$
 - $m_I = \pm 3/2, \pm 1/2$
- $$H_{\text{hf}} = A I \cdot J + P I_z^2$$



Outline

Di Vincenzo 1996



Isolation



Read-out



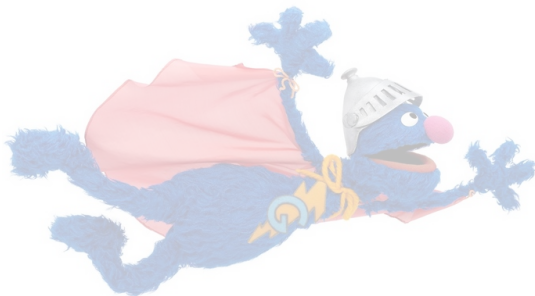
Initialization



Manipulation



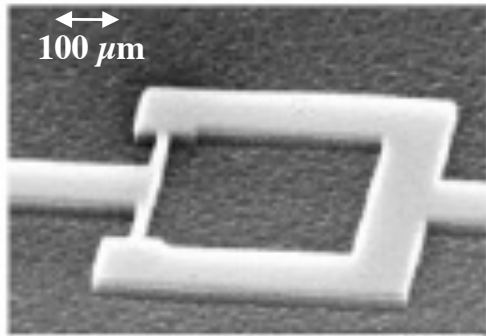
Storage



Quantum gates, Grover's algorithm

SMM detectors

- tiny magnetic moment (few μ_B) \rightarrow sensitive detectors

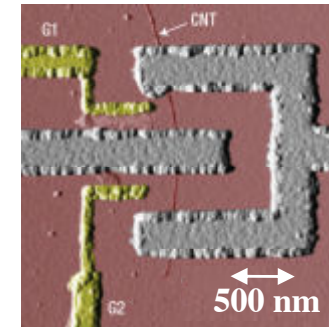


Phys. Rev. Lett. **77**, 1873 (1996)

sensitivity :
 $10^2 - 10^3$ spins

size reduction \rightarrow

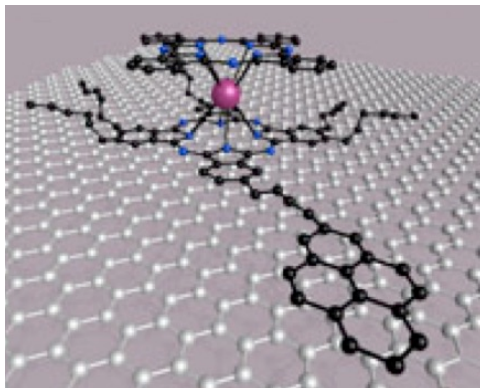
Nano-SQUID



Nature Nano **1**, 53 (2006)

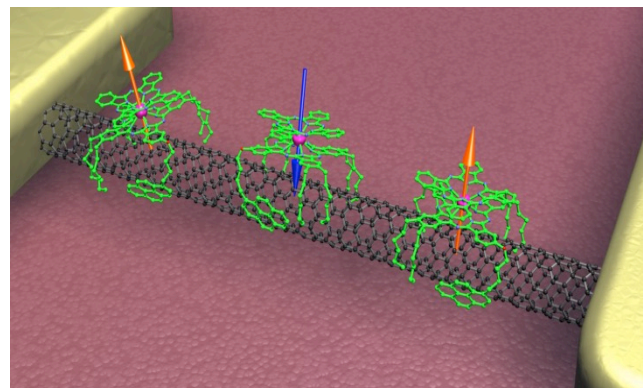
\downarrow different concepts

2D



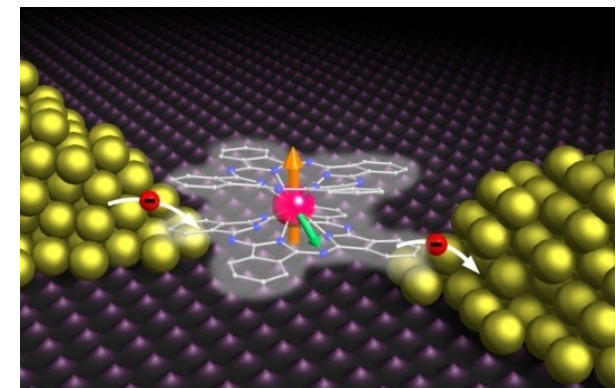
Nano Lett. **11**, 2634 (2011)

1D



Nature Materials **10**, 502 (2011)

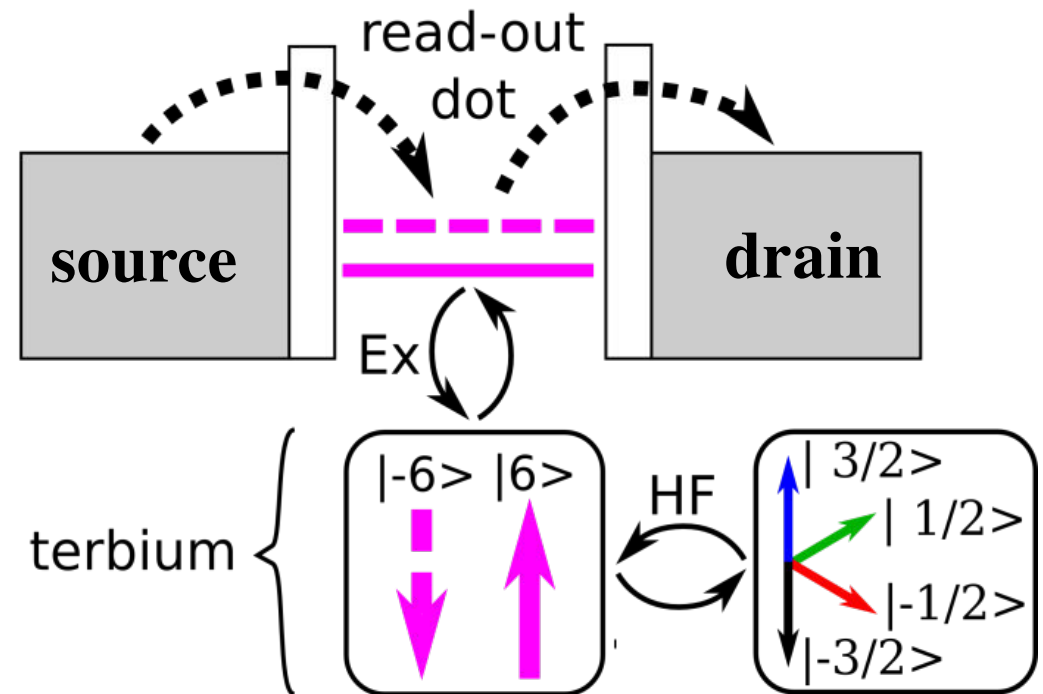
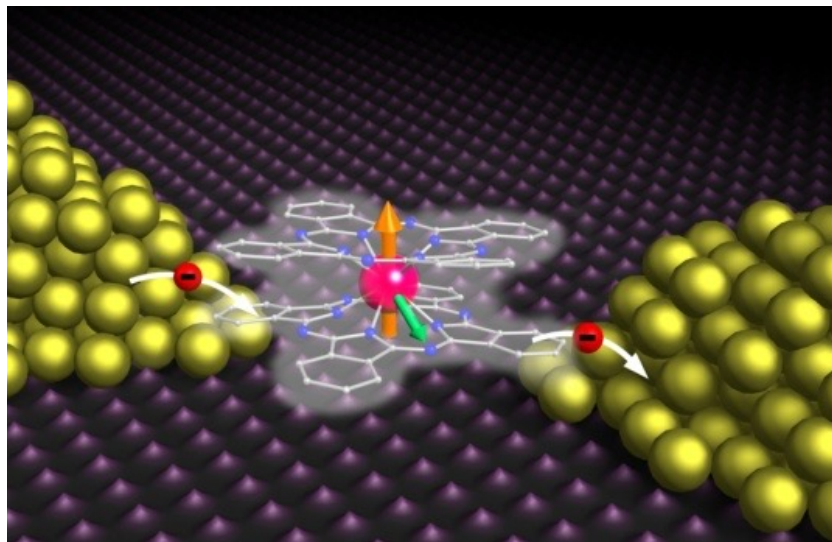
0D



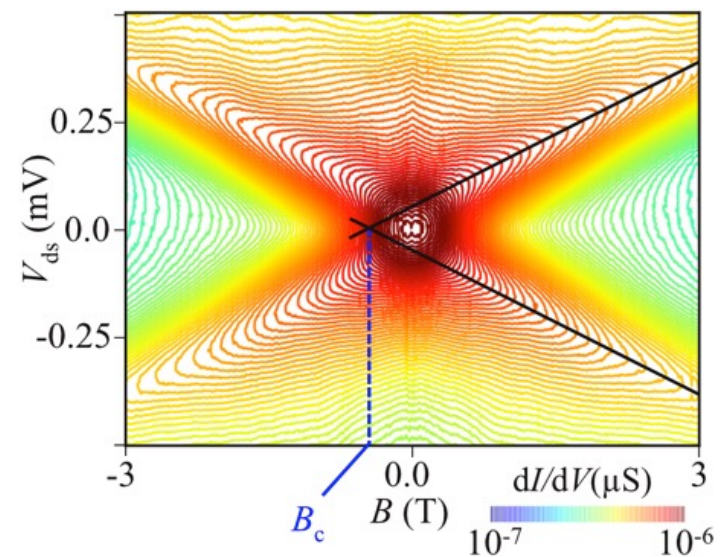
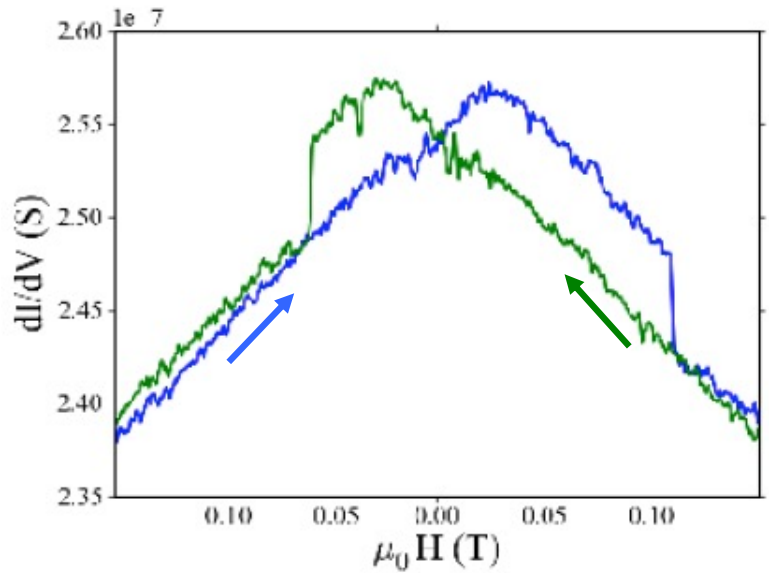
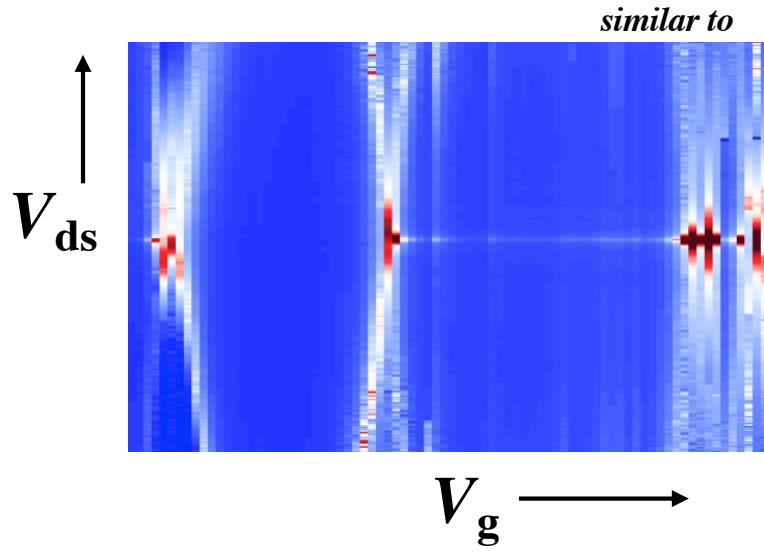
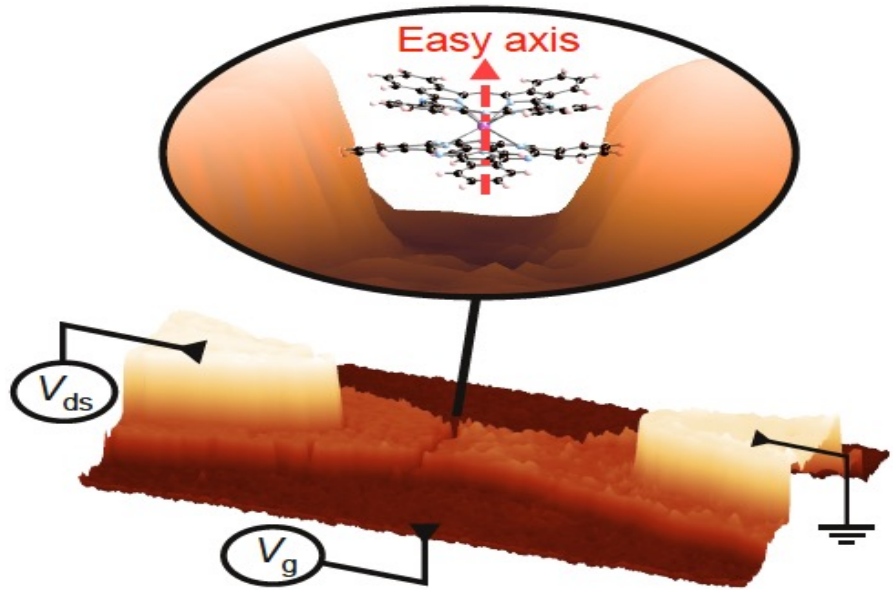
Nature **488**, 357 (2012)

Schematics of the read-out

Weak two-stage-coupling to avoid backaction

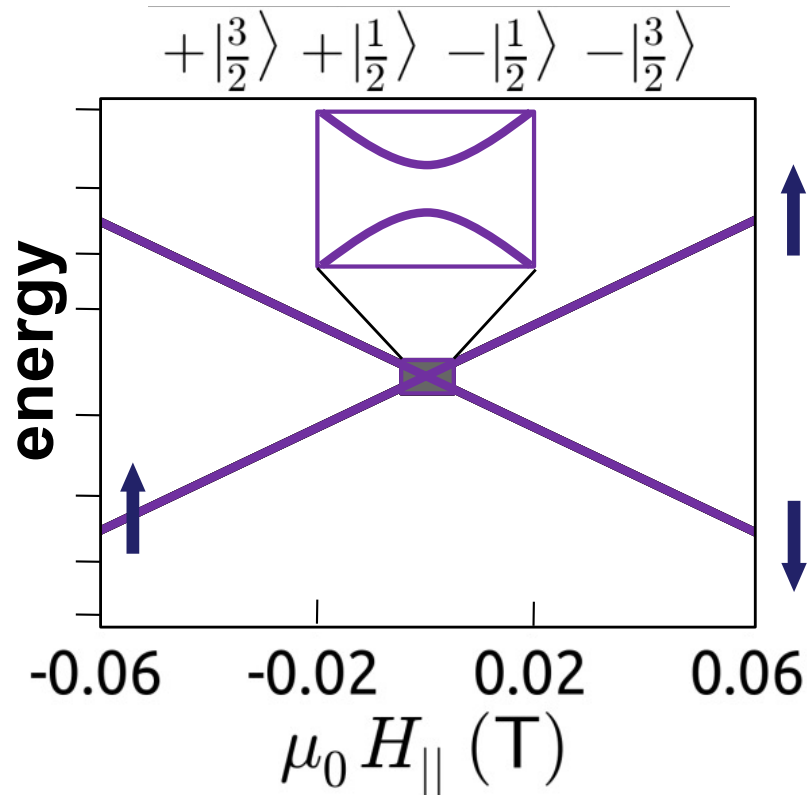
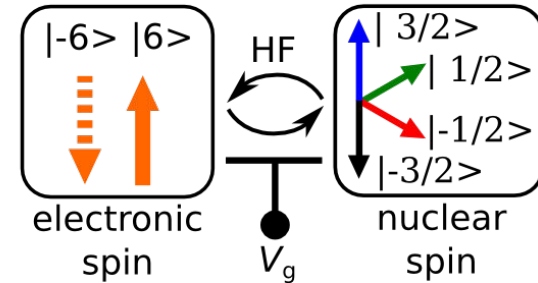


Device characteristics



Nuclear spin read-out

- ligand field induces anti-crossings



at each anticrossing:

- e⁻ spin reversal
- nuclear spin conservation

position of e⁻ spin reversal

linked to nuclear spin state



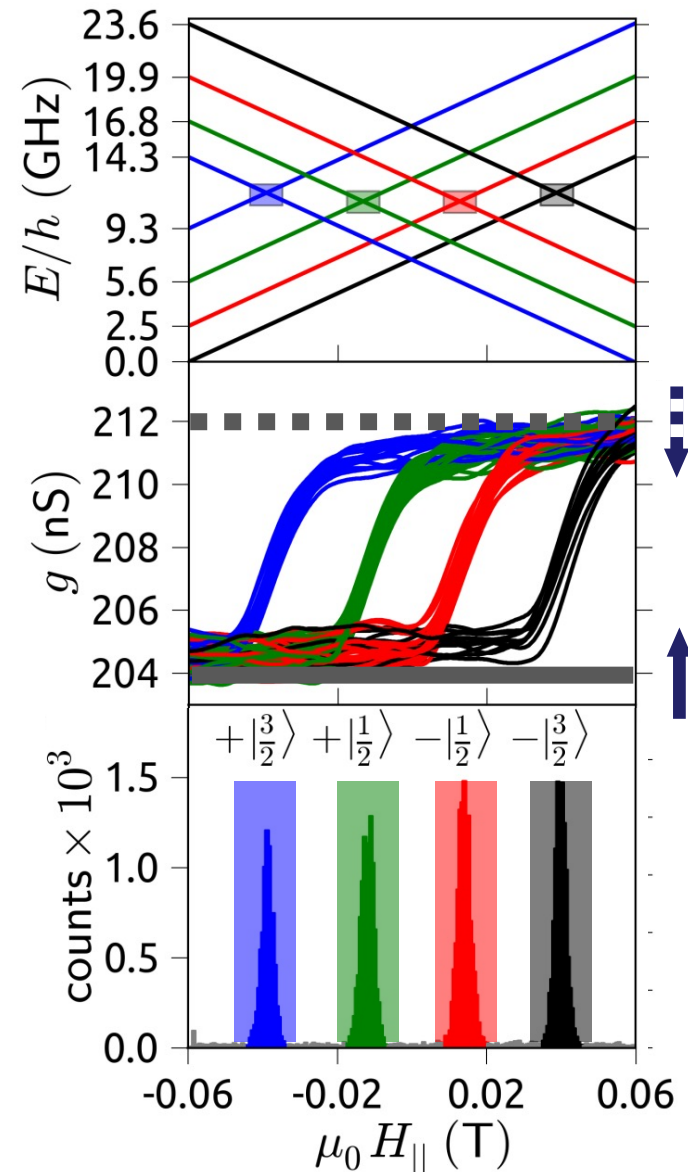
Nuclear spin read-out

1st nuclear spin → electronic spin

2nd electronic spin → read-out dot

- overlap between Tb e⁻ wavefunction conjugated pi-orbitals of the Pc → exchange interaction
- coupling of e⁻-spin to tunnel current
- spin reversal = conductance jump
- ramping the magnetic field (75000x) & recording conductance jumps

conduction jump position yields nuclear spin state



Outline

Di Vincenzo 1996



Isolation



Read-out



Initialization



Manipulation



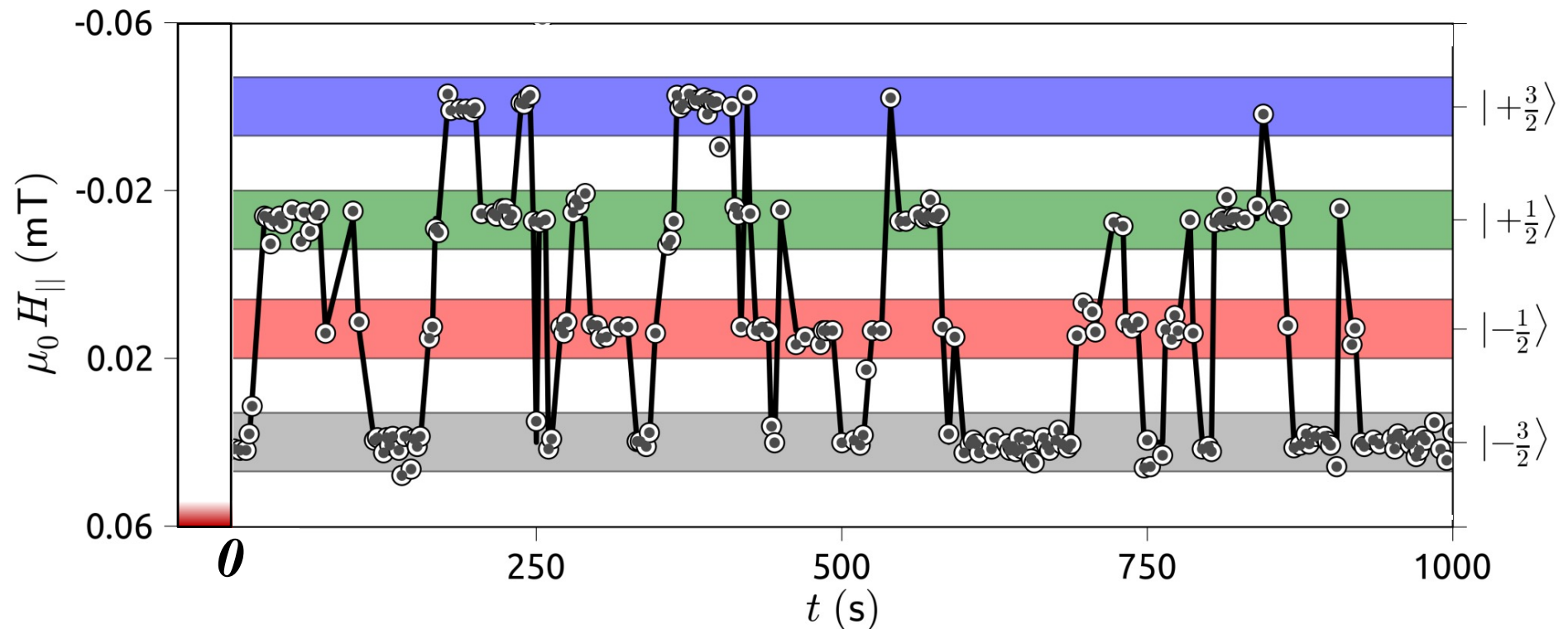
Storage



Quantum gates, Grover's algorithm

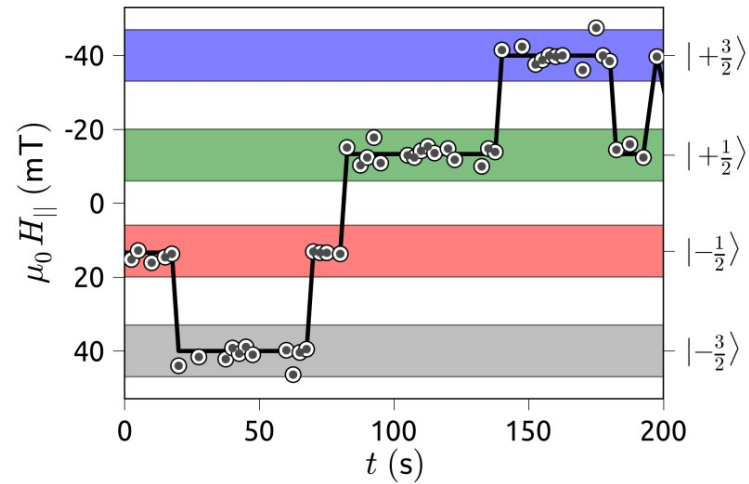
Nuclear spin trajectory

- sweeping the magnetic field back and forth
- each jump \rightarrow nuclear spin state + time



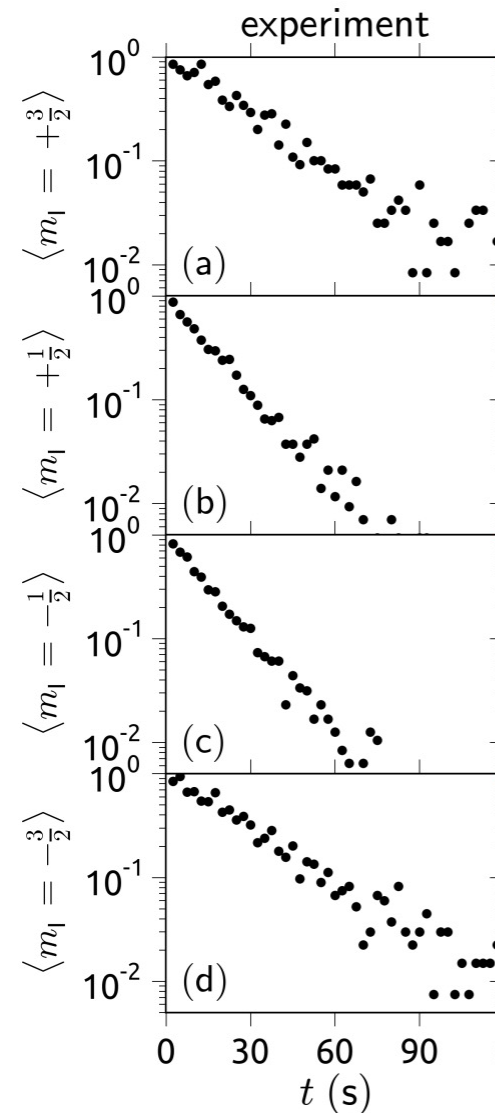
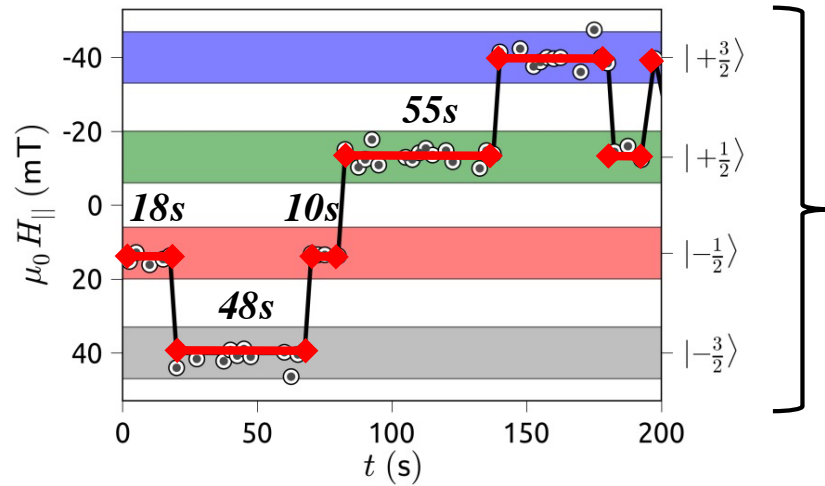
Thiele et al. PRL 111, 3 (2013)

Single nuclear spin relaxation



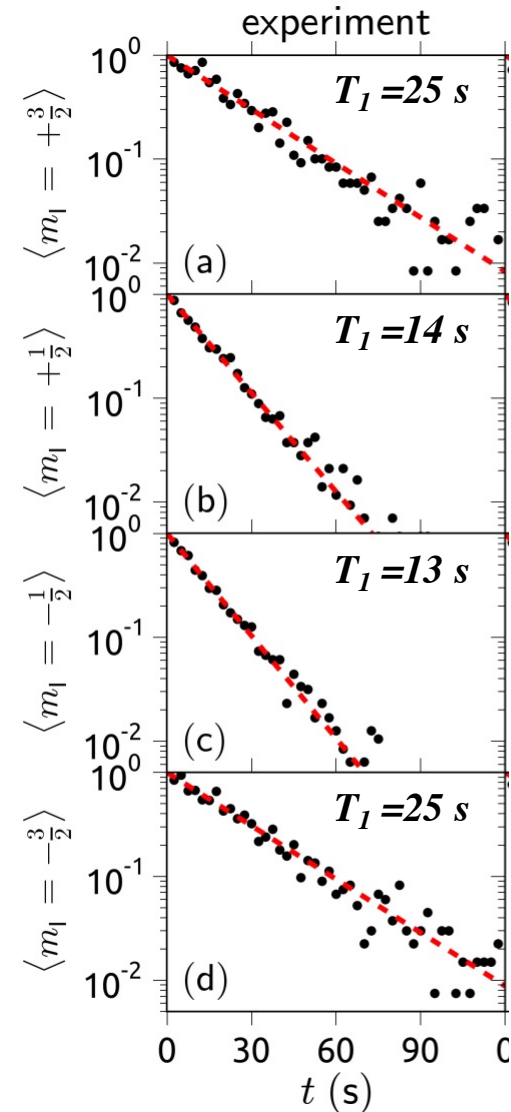
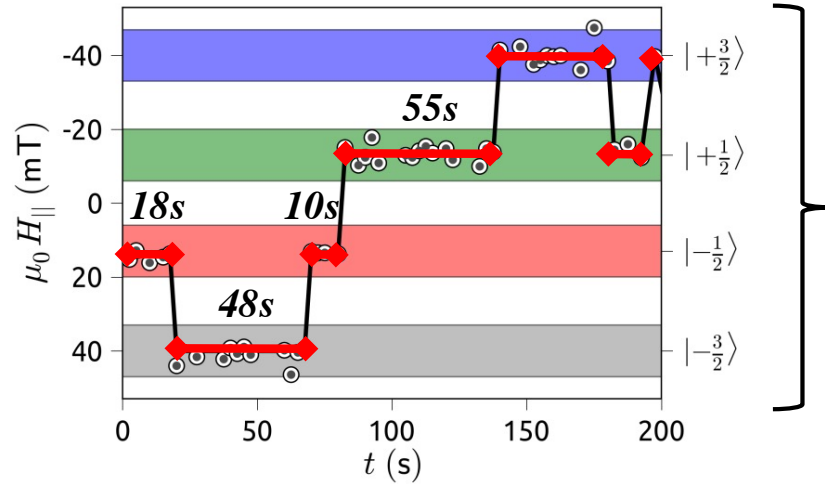
Thiele et al. PRL 111, 3 (2013)

Single nuclear spin relaxation



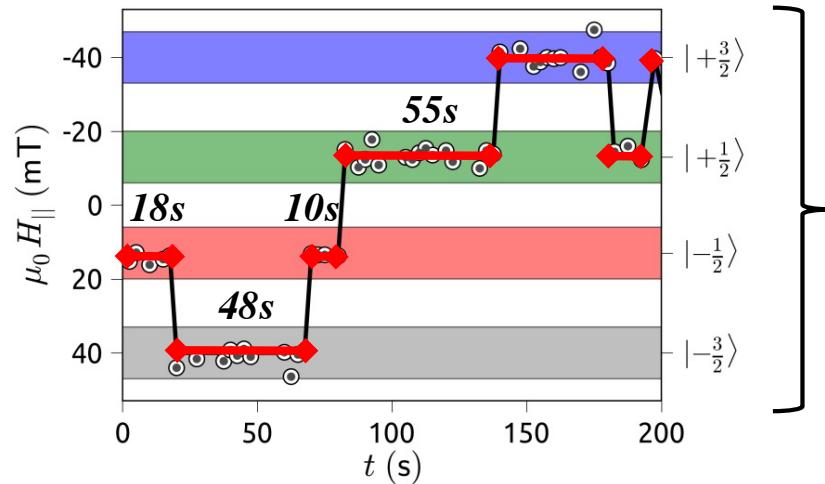
Thiele et al. PRL 111, 3 (2013)

Single nuclear spin relaxation



Thiele et al. PRL 111, 3 (2013)

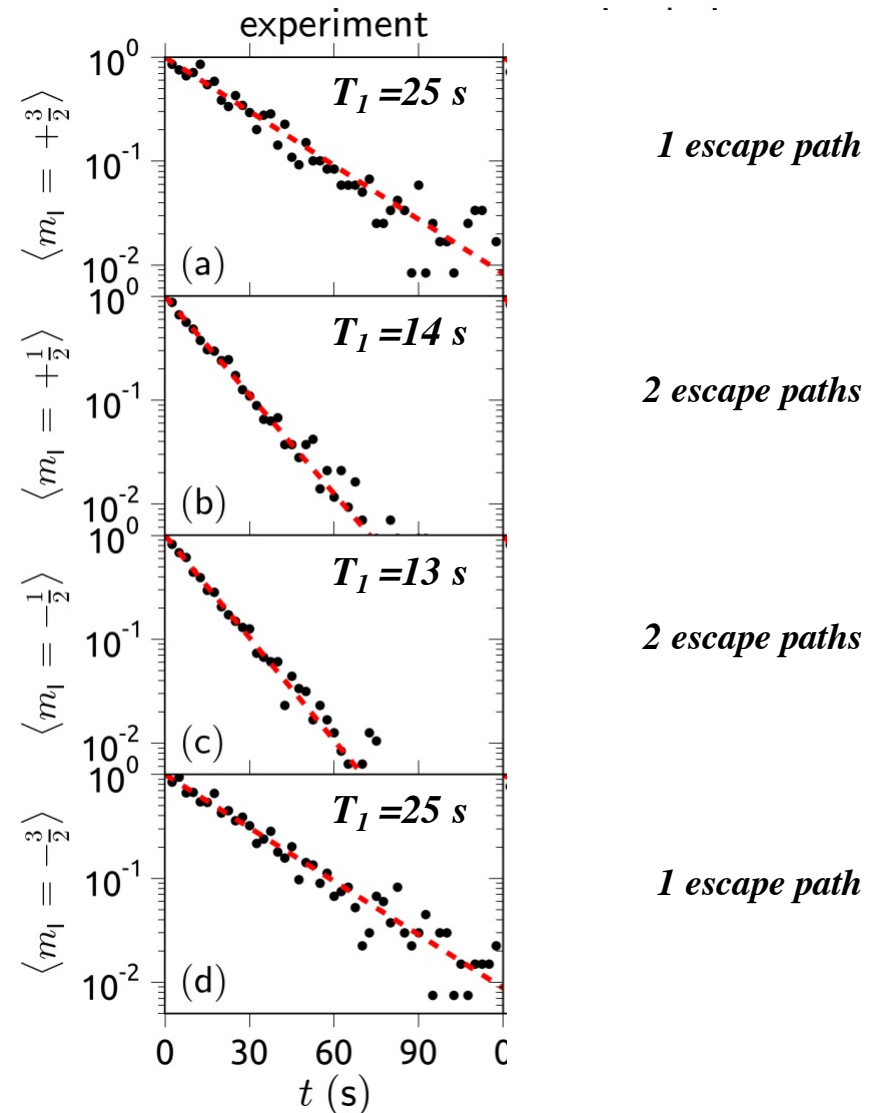
Single nuclear spin relaxation



- plot dwell times in a histogram
- fit to $e^{-t/T_1} \rightarrow$ relaxation time $T_1 \approx 10$ s
- difference of 2 in $T_1 \rightarrow$ relaxation path

\rightarrow measurement = initialization

Thiele et al. PRL 111, 3 (2013)



Outline

Di Vincenzo 1996



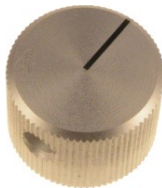
Isolation



Read-out



Initialization



Manipulation



Storage



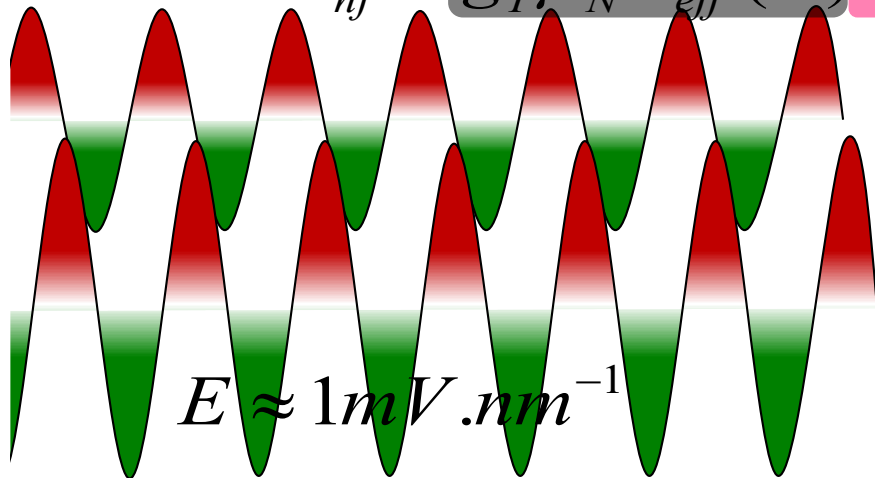
Quantum gates, Grover's algorithm

Single Nuclear Spin Manipulation

$$H_{hf} = [A_0 + \Delta A(E)] J \cdot I + P I_z^2$$

Electric field!
Magnetic field

$$= H_{hf} + g_I \mu_N B_{eff}(E) I$$



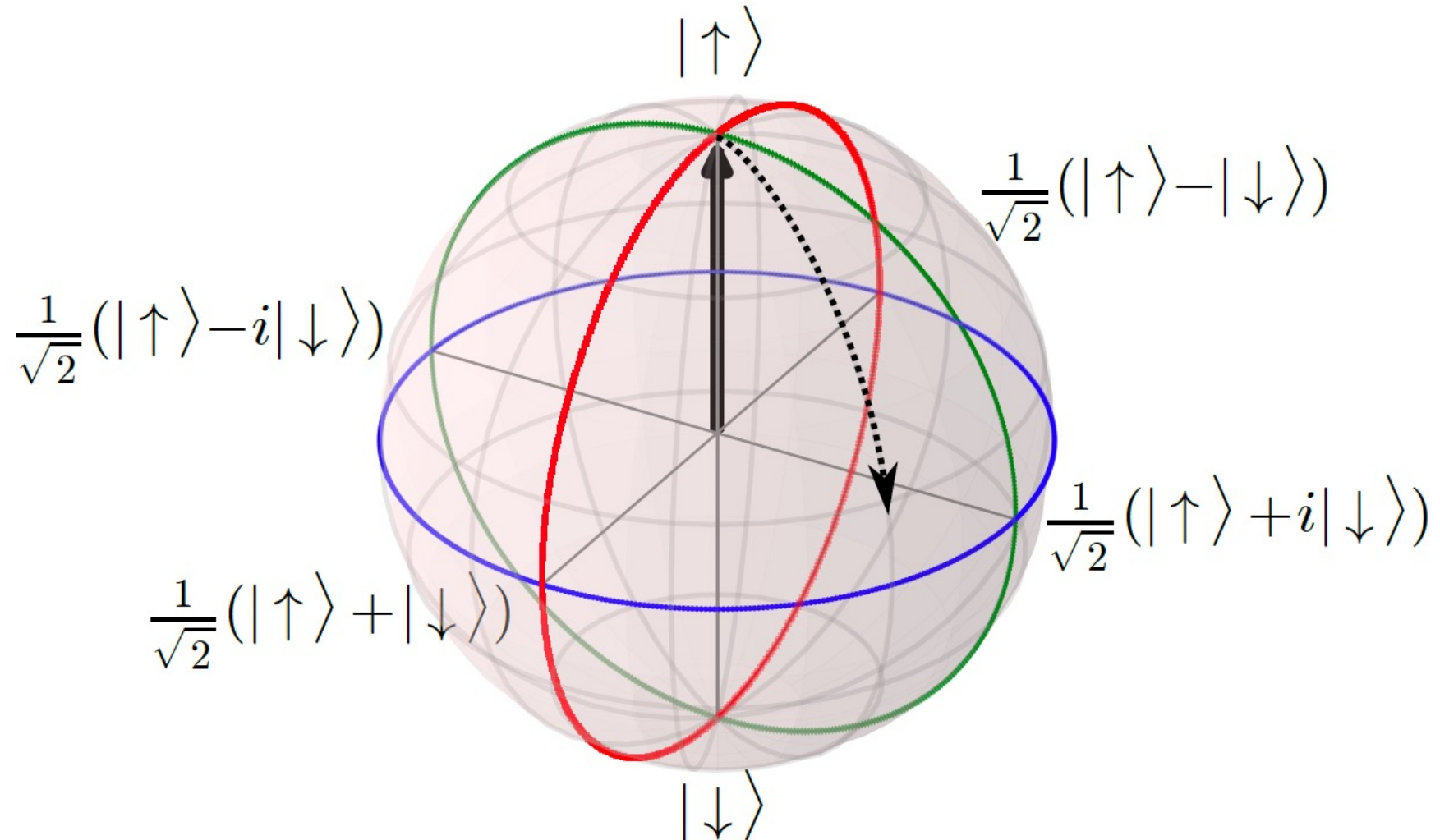
$$\frac{\Delta A}{A_0} \approx \frac{1}{10000}$$

$$B_{eff} \approx 100 mT$$

Hyperfine interaction



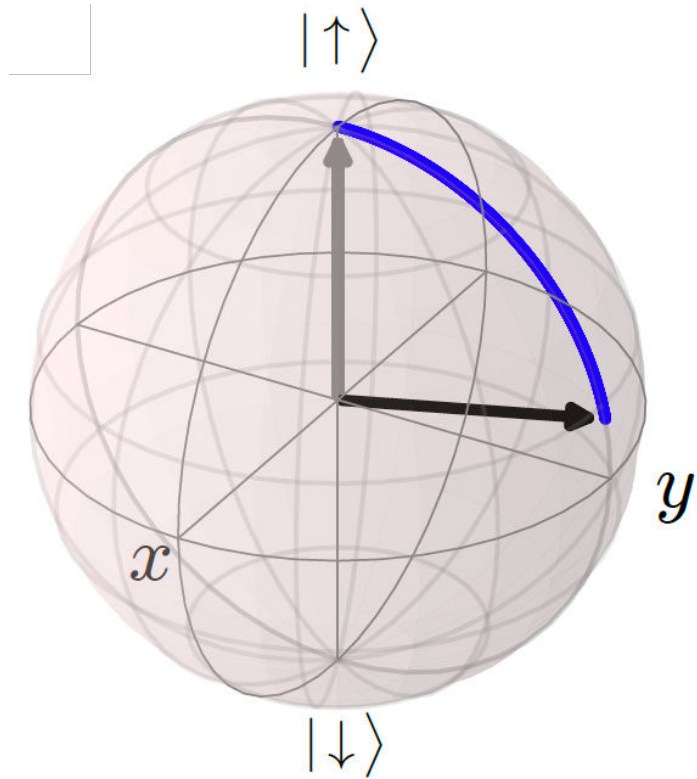
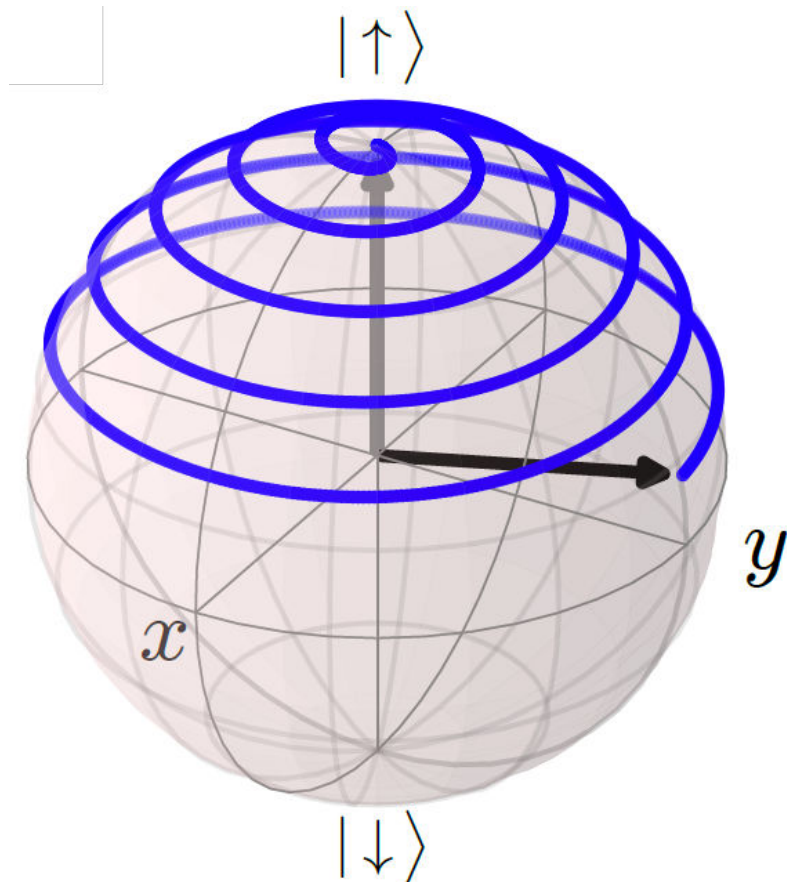
Bloch sphere representation of a two level spin qubit system



Trajectory of a spin qubit, in the laboratory frame and the rotating frame

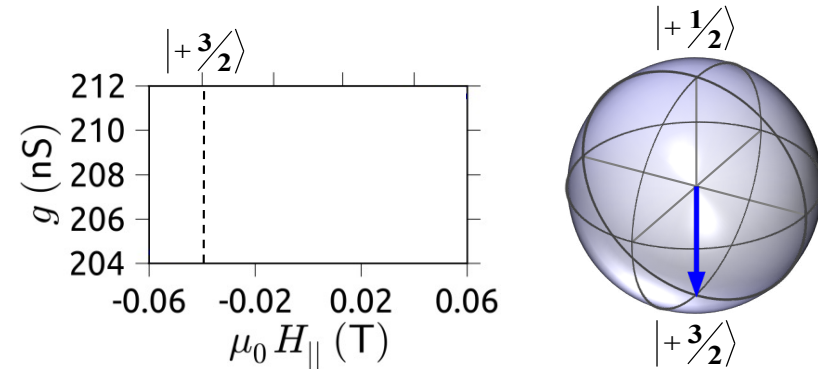
$$H = \hbar\omega_z\sigma_z + \hbar\Omega (\sigma_x\cos(\omega t) + \sigma_y\sin(\omega t))$$

$$H = \frac{\hbar\Delta}{2}\sigma_z + \frac{\hbar\Omega}{2}\sigma_x$$



Nuclear spin manipulation

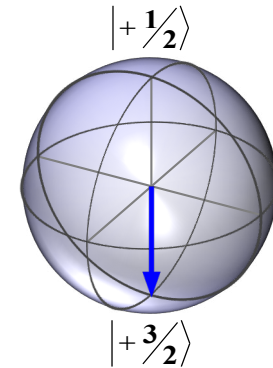
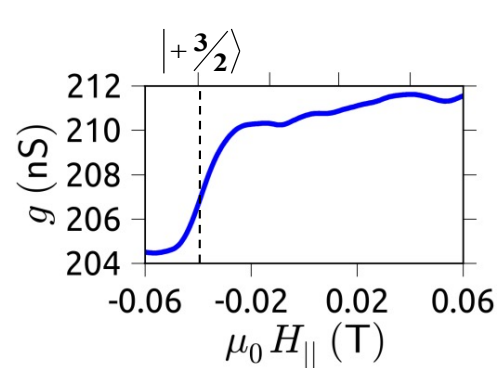
- I. **Initialization:**
sweeping the external mag. field +
position of the conductance jump
initializes the spin



Nuclear spin manipulation

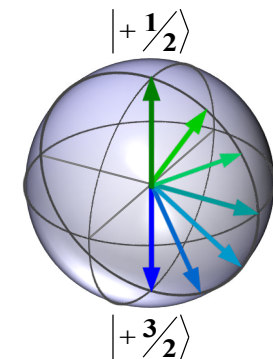
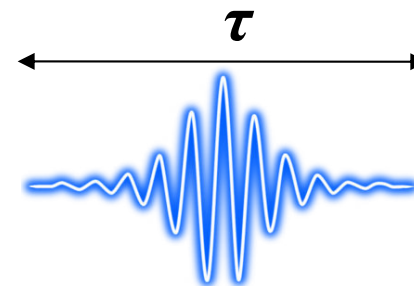
I. Initialization:

sweeping the external mag. field +
position of the conductance jump
initializes the spin



II. Pulse:

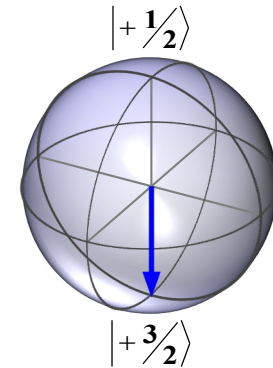
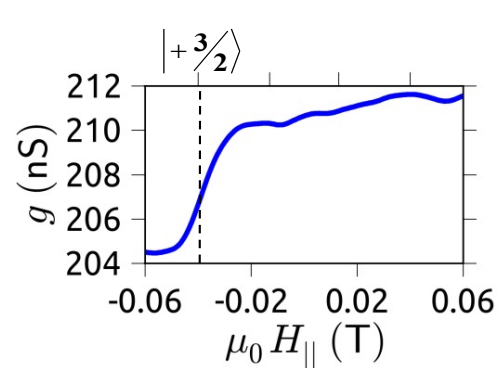
applying an oscillating electric field
with the frequency f and different
durations τ creates a coherent
superposition



Nuclear spin manipulation

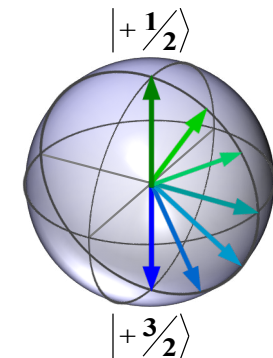
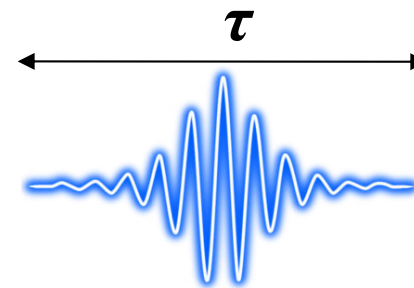
I. Initialization:

sweeping the external mag. field +
position of the conductance jump
initializes the spin



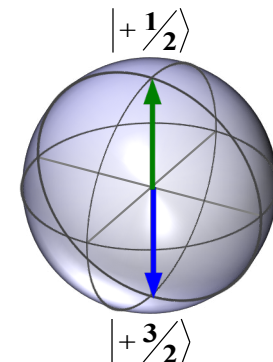
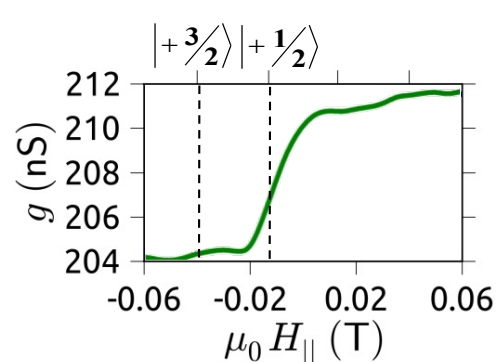
II. Pulse:

applying an oscillating electric field
with the frequency f and different
durations τ creates a coherent
superposition



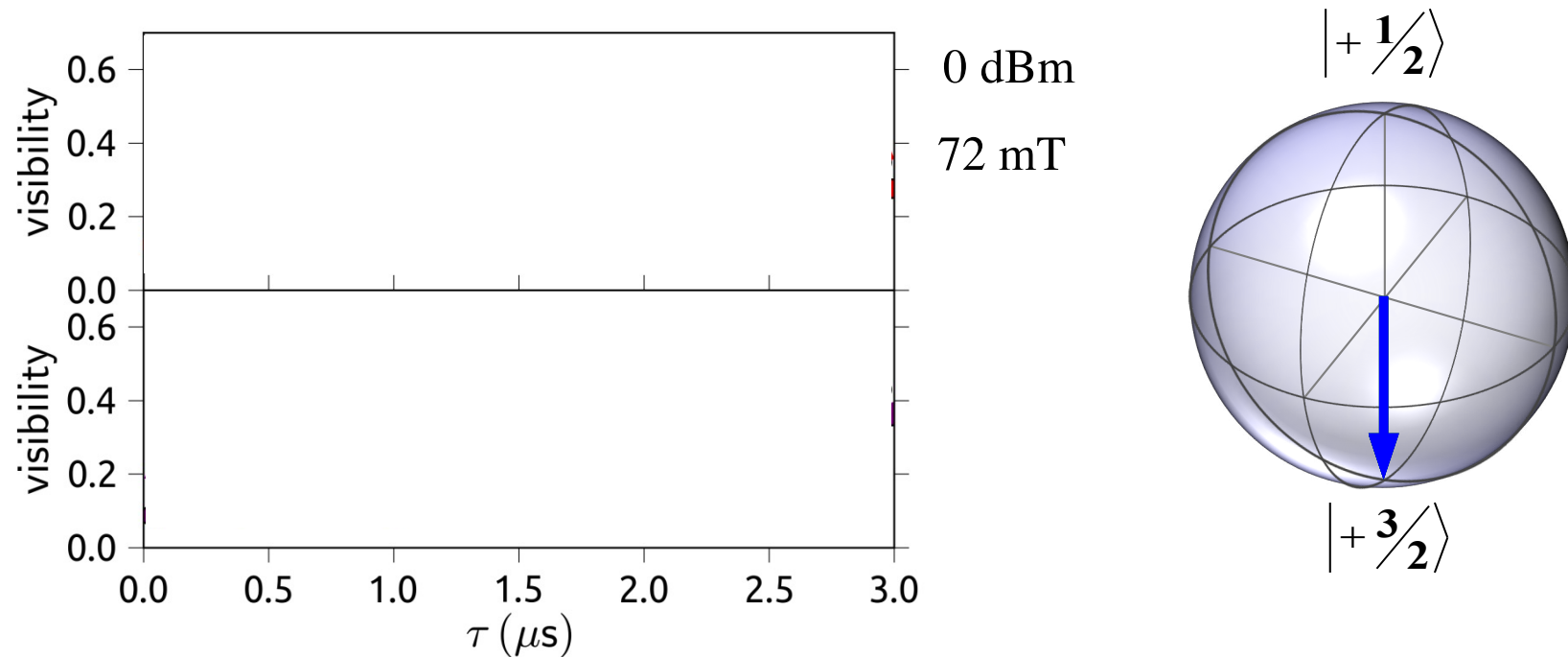
III. Read-out:

sweeping the external mag. field +
position of the conductance jump
projects the spin



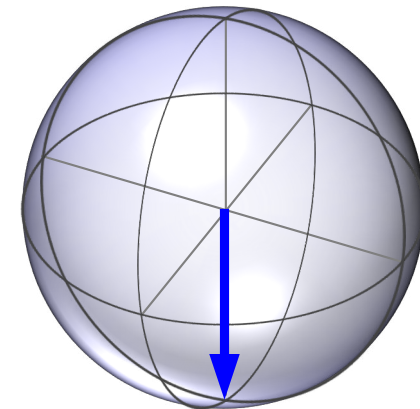
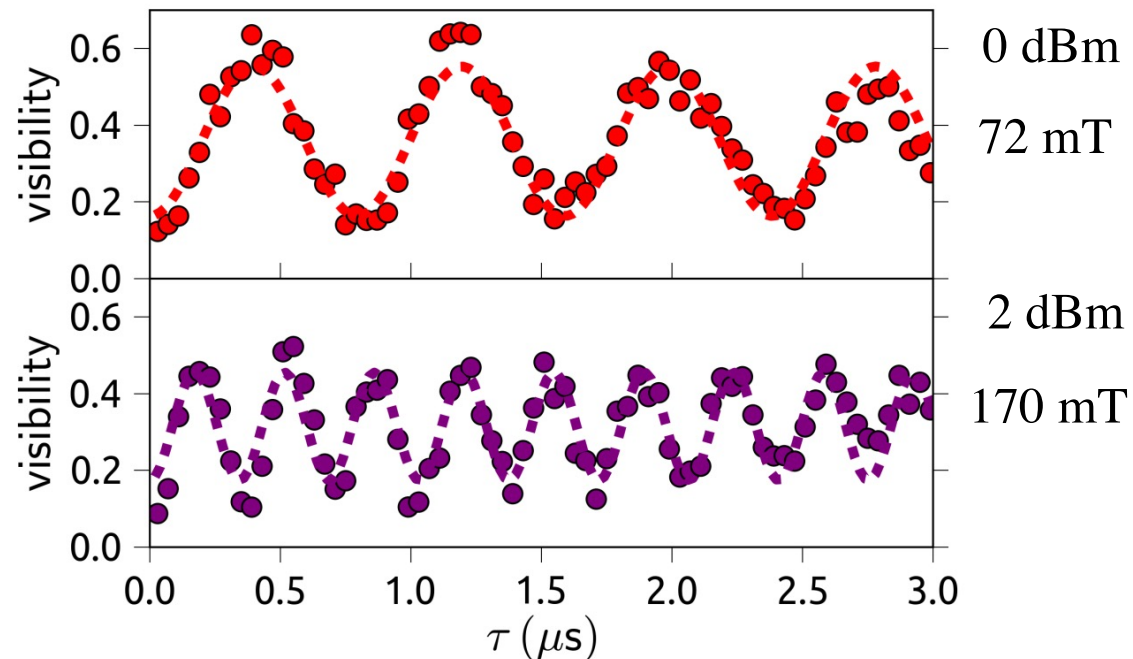
Nuclear spin manipulation

- repeating this procedure 100x for different pulse durations and two different microwave powers
- plotting the excited state probability



Nuclear spin manipulation

- repeating this procedure 100x for different pulse durations and two different microwave powers
- plotting the probability of staying in the state



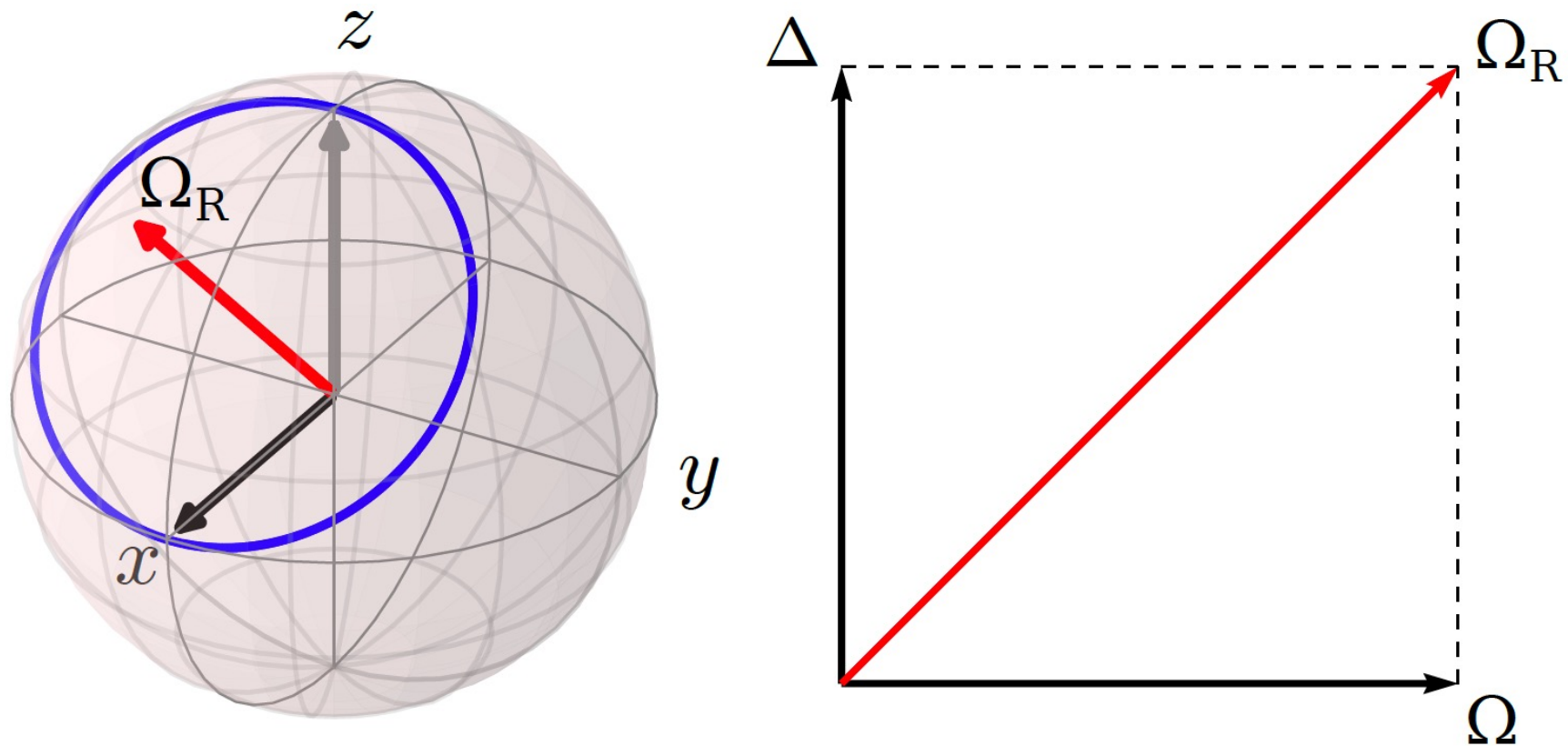
- Rabi oscillation: direct proof of coherent superposition + manipulation
- created using the AC Stark effect

S. Thiele et al., Science 344,

1135 (2014)

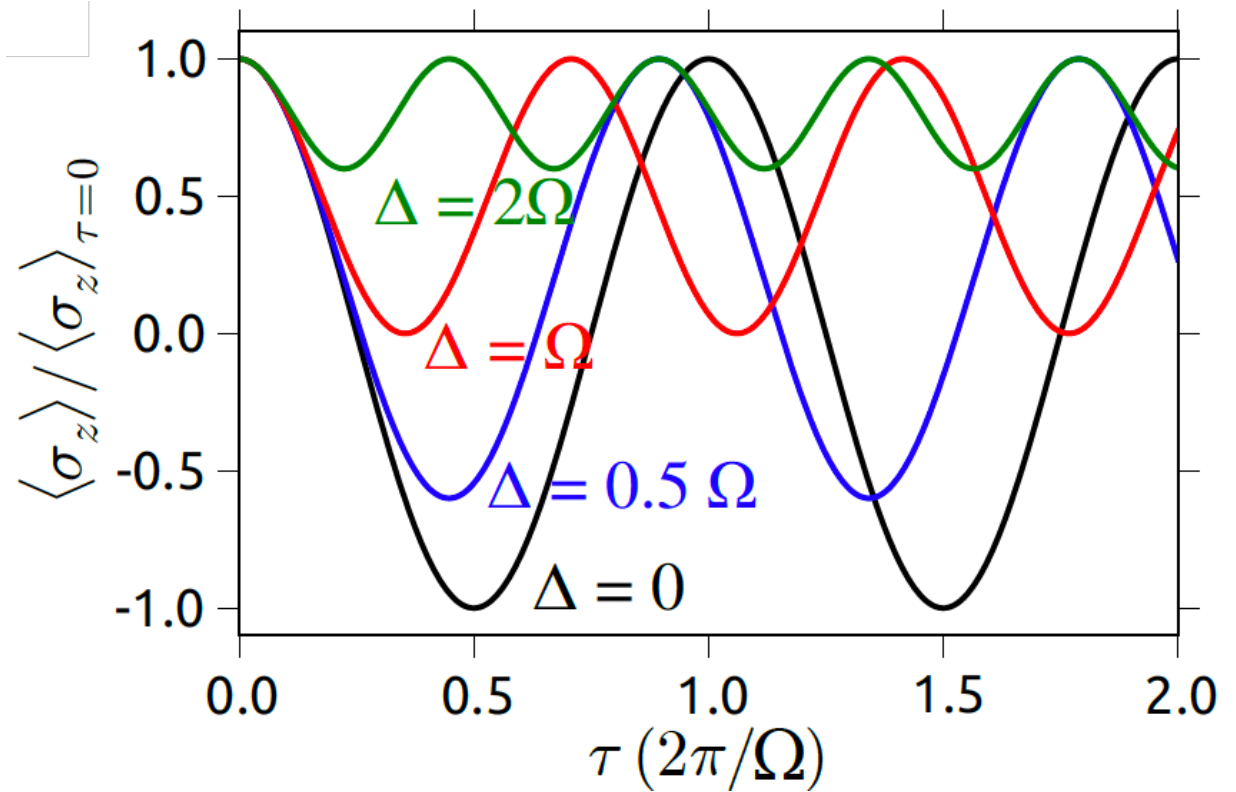
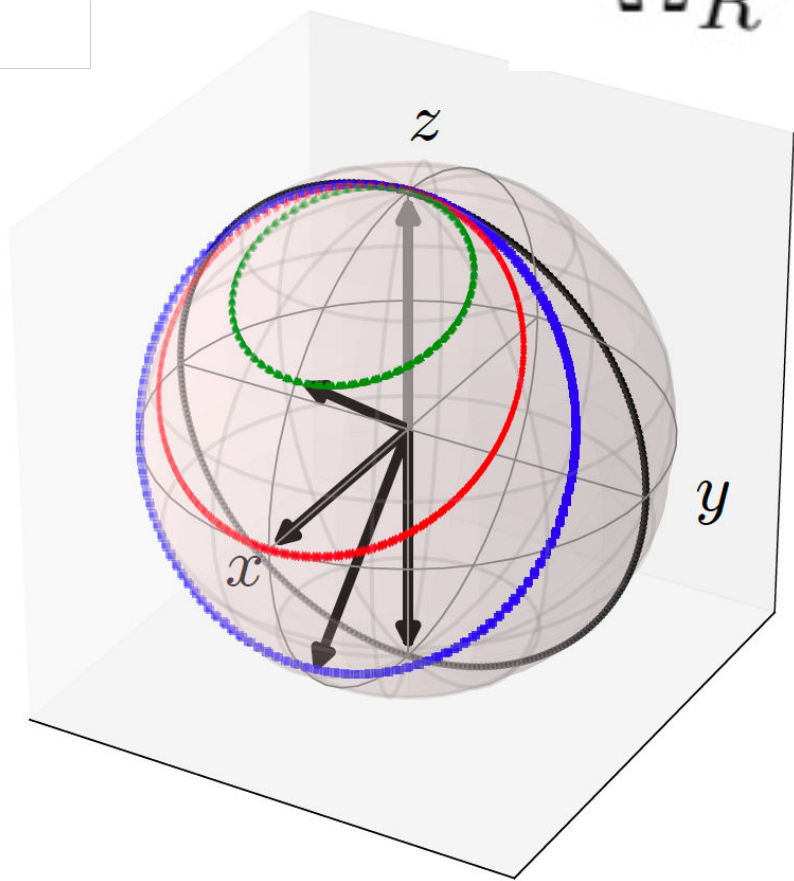
Spin precession around an effective magnetic field in the rotating frame

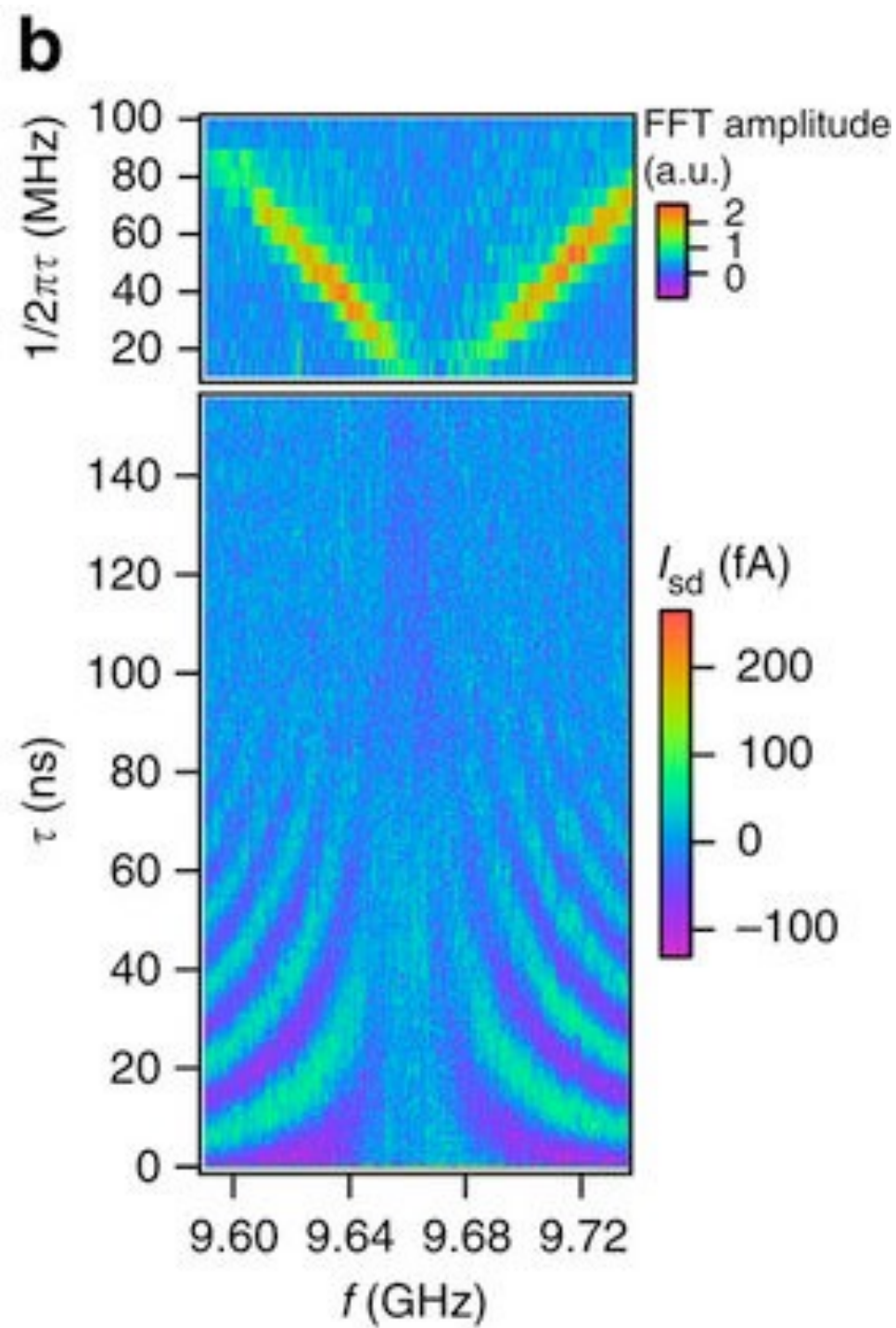
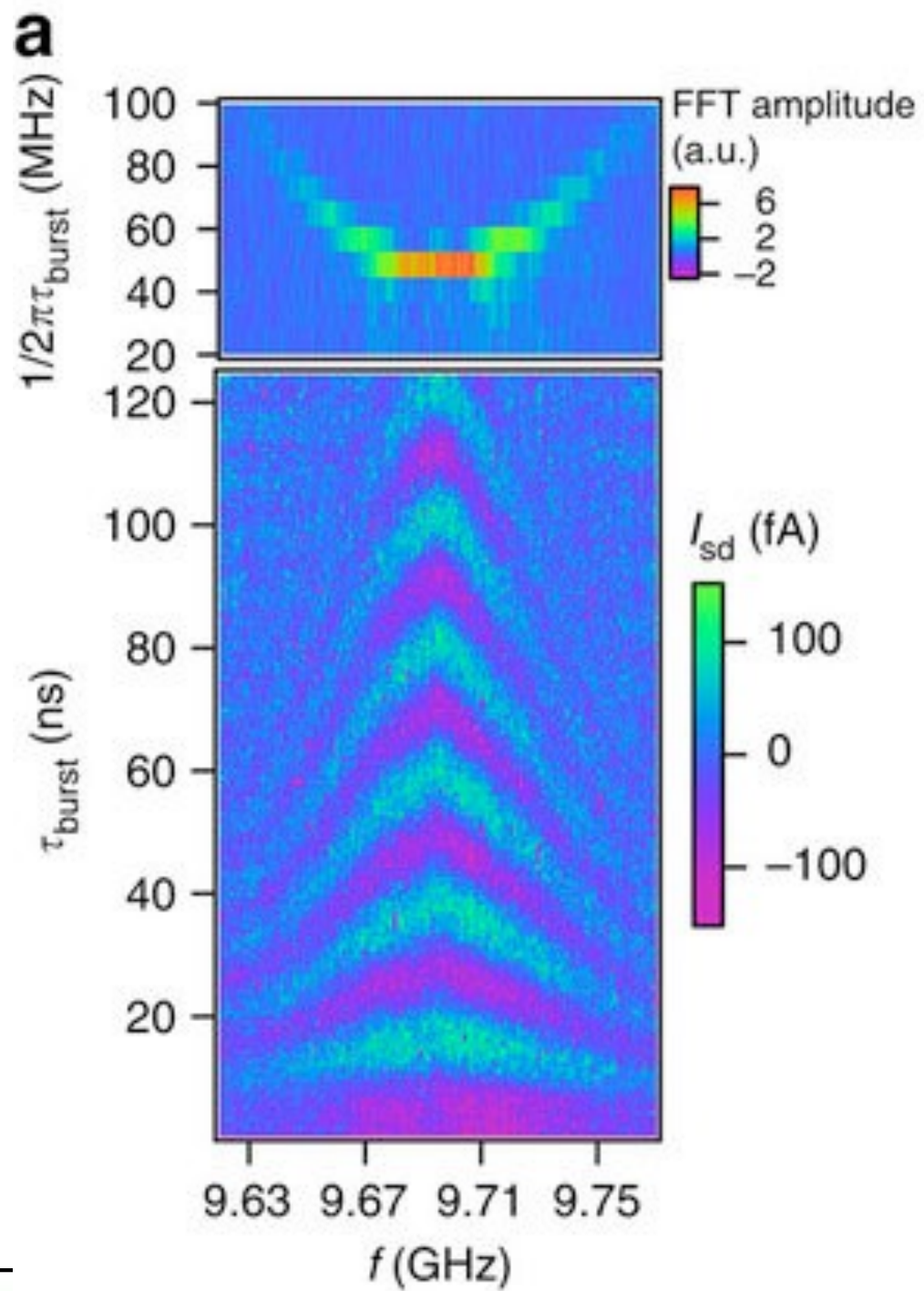
$$\Omega_R = \sqrt{\Delta^2 + \Omega^2}$$

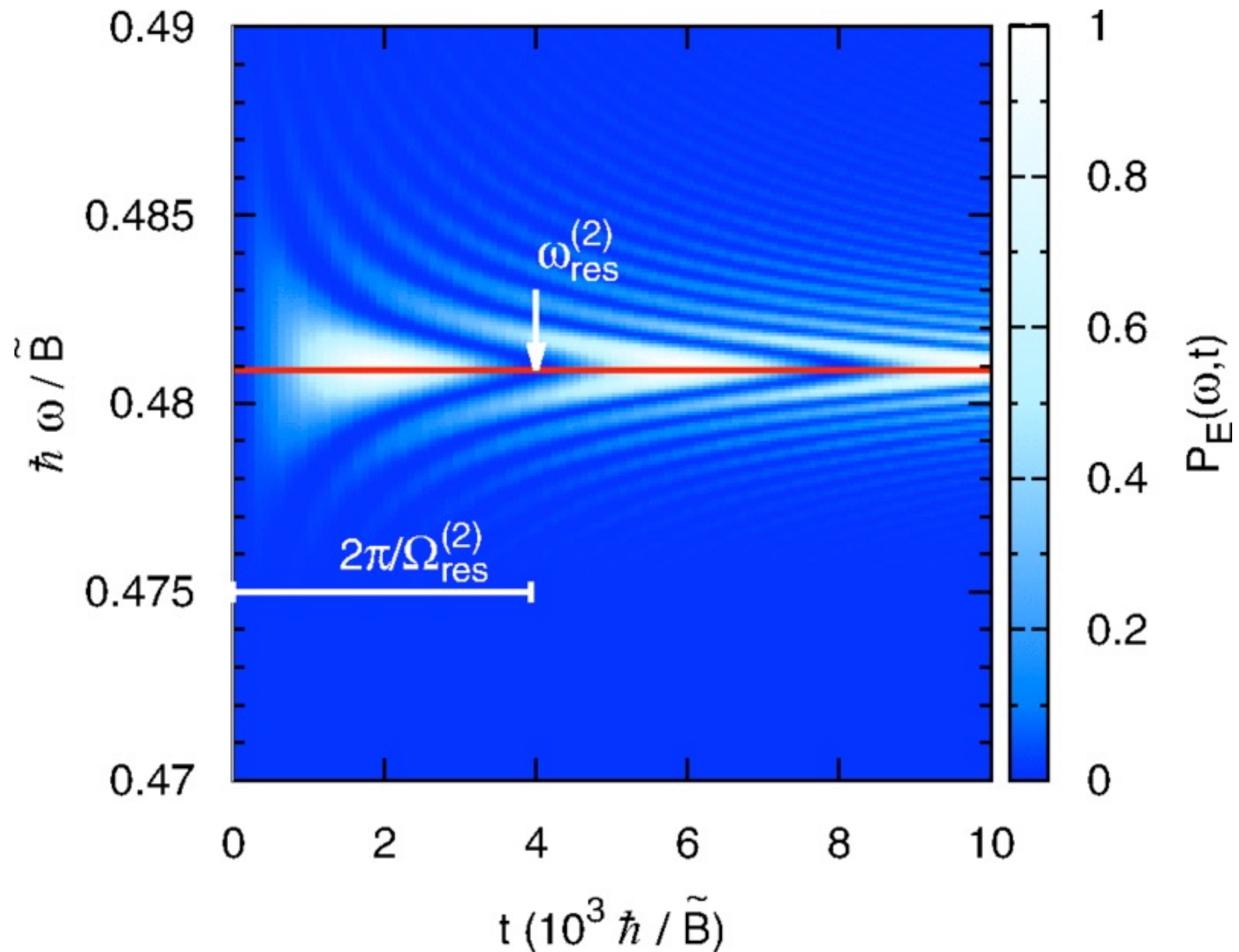


Trajectory of the Bloch vector in the rotating frame at different detunings

$$\Omega_R = \sqrt{\Delta^2 + \Omega^2}$$





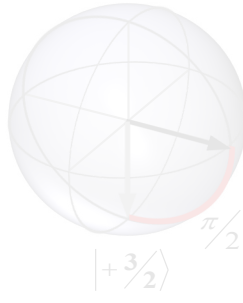


Basic quantum operation: Ramsey

1 - initialization



2 - coherent superposition



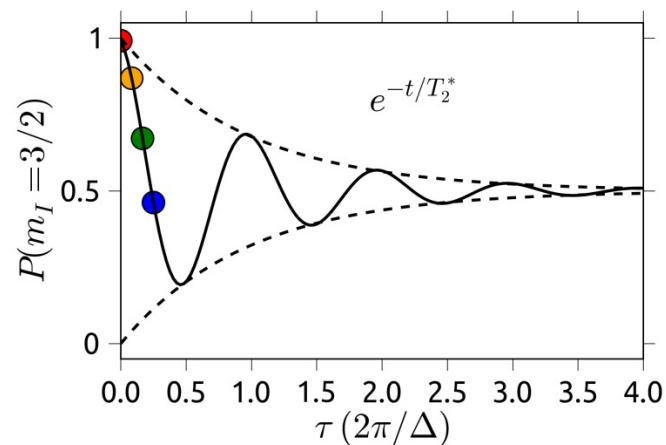
3 - free evolution



4 - rotation

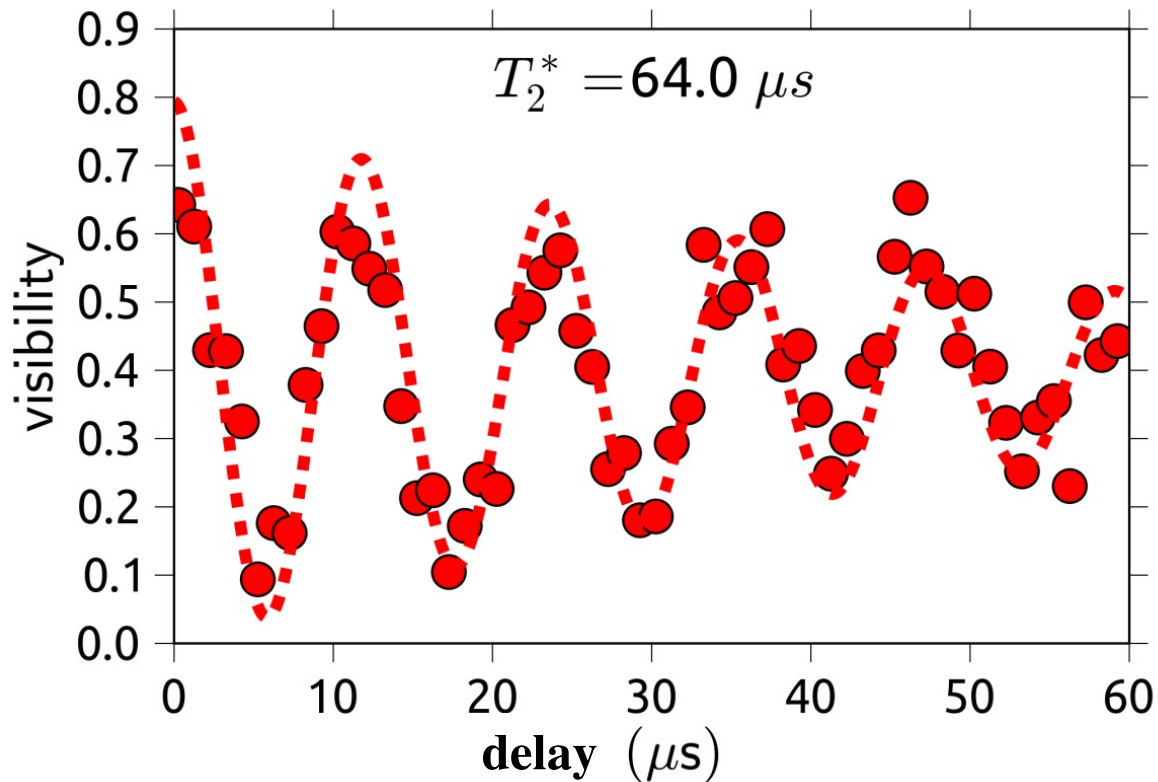
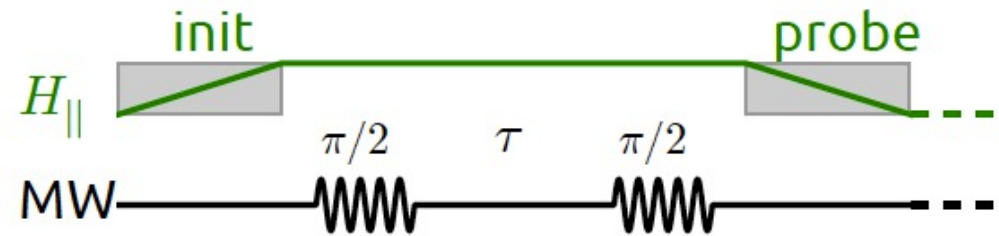


5 - projection



- Oscillation : Vector rotation
- Damping : Decoherence

Basic quantum operation: Ramsey



*S. Thiele et al., Science 344,
1135 (2014)*

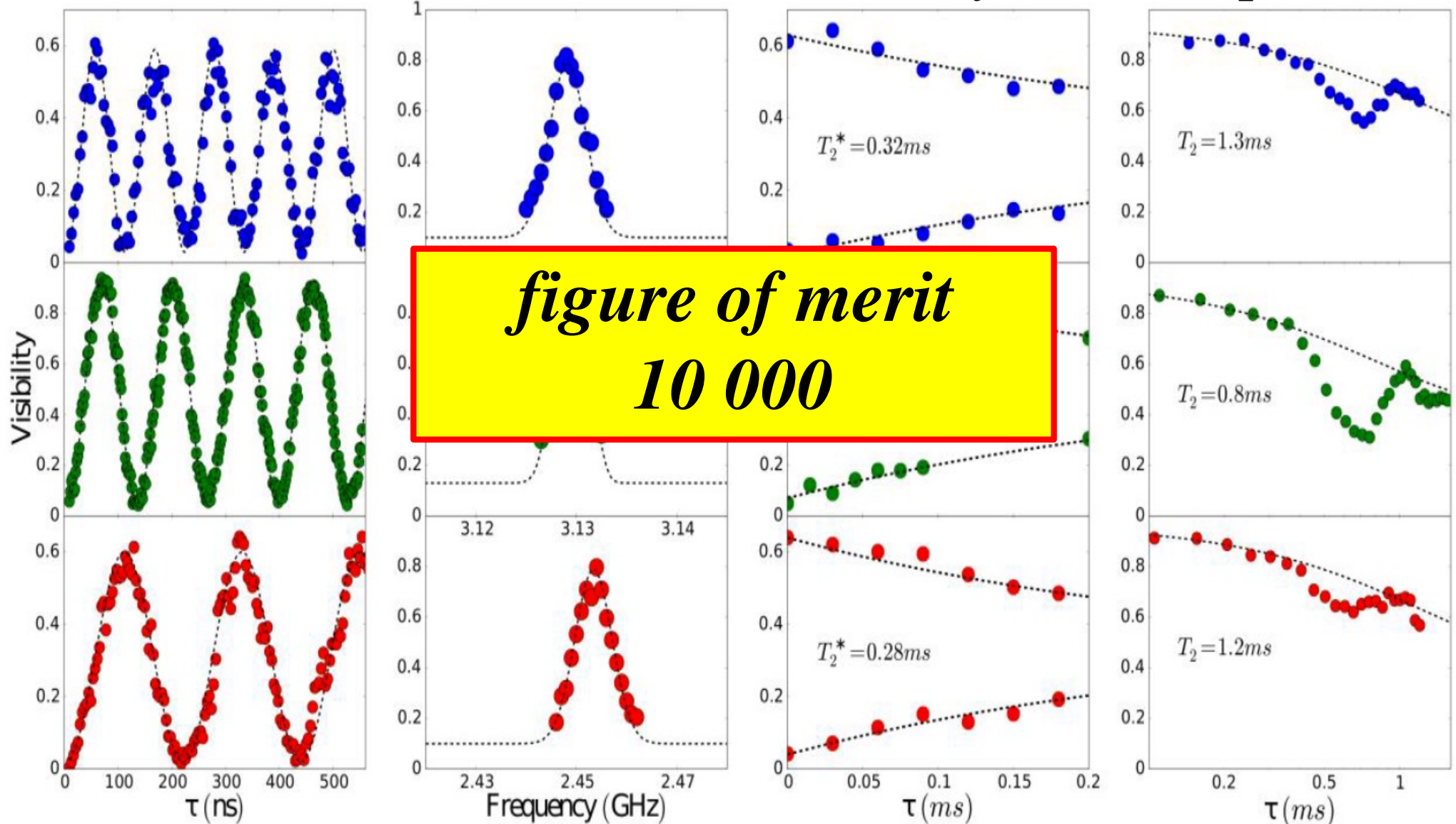
Basic quantum operation

Rabi

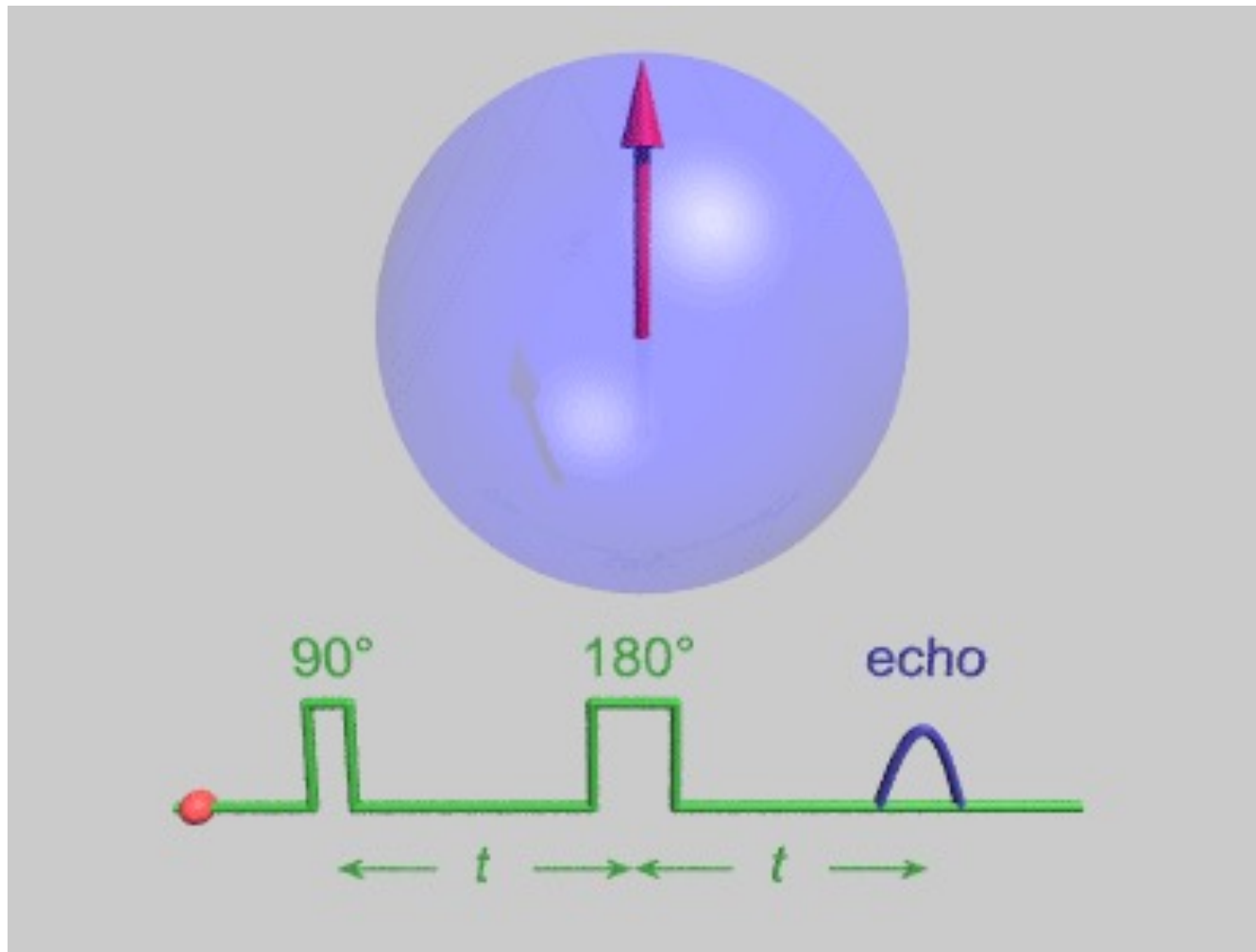
Resonance

Ramsey

Hahn spin echo

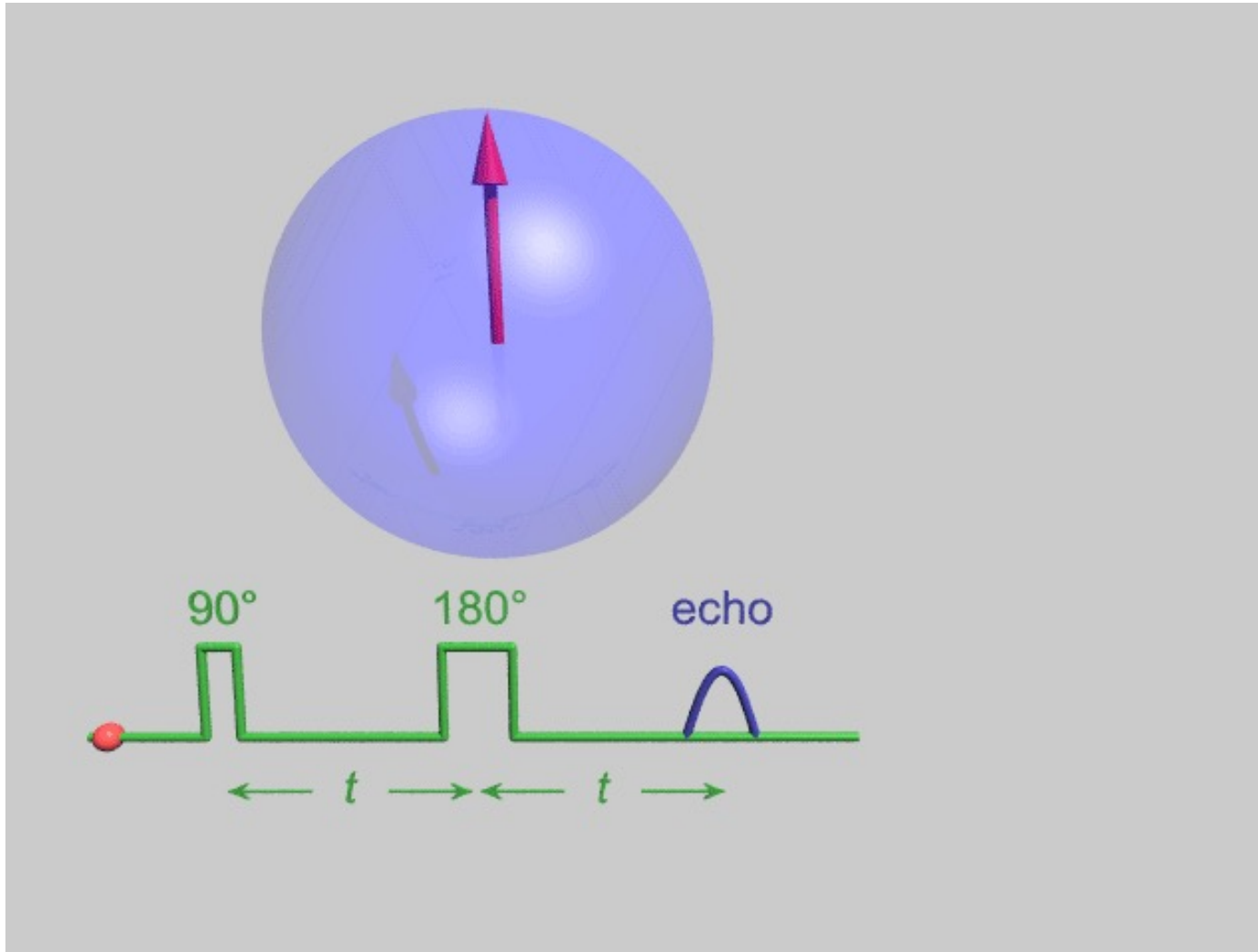


Hahn Spin Echo



*By Gavin W Morley - Own work, CC BY-SA 3.0,
<https://commons.wikimedia.org/w/index.php?curid=15230576>*

Hahn Spin Echo Decay



*By Gavin W Morley - Own work, CC BY-SA 3.0,
<https://commons.wikimedia.org/w/index.php?curid=15230576>*

Outline

Di Vincenzo 1996



Isolation



Read-out



Initialization



Manipulation



Storage



Quantum gates, Grover's algorithm

Outline

Di Vincenzo 1996



Isolation



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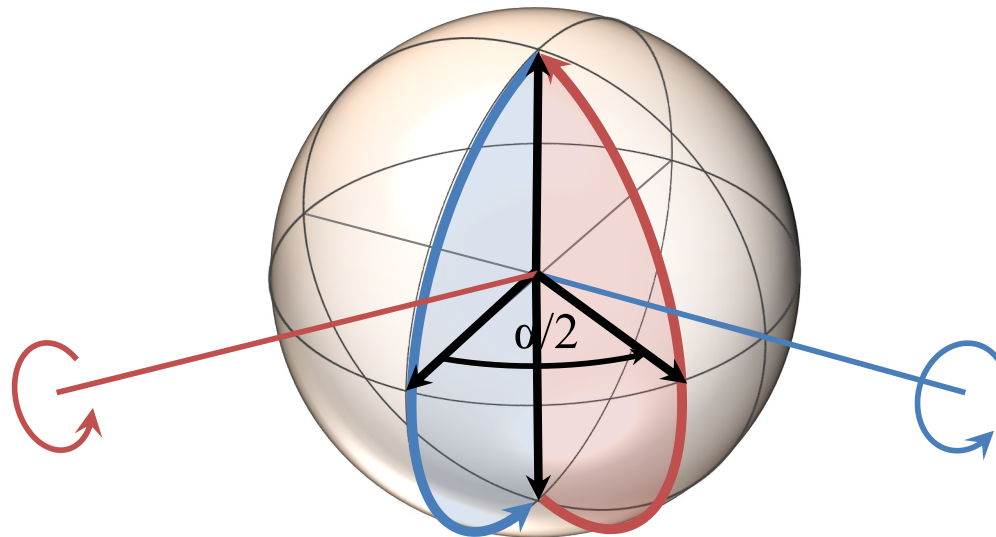


Quantum gates, Grover's algorithm

Geometric phase (Berry's phase)

Geometric phase accumulation :

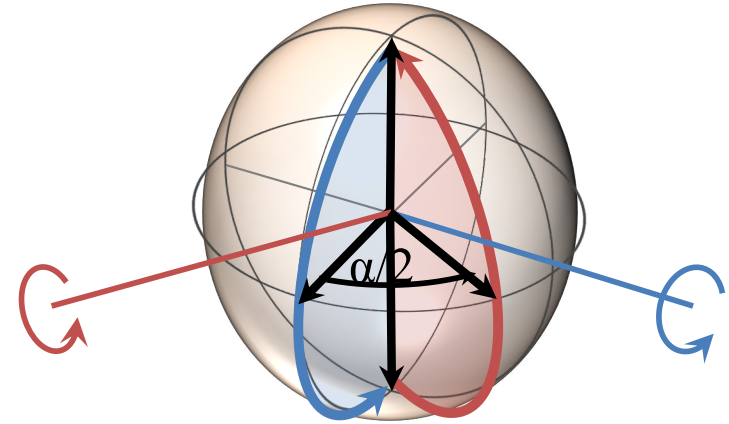
- 1) *Pulse π with $\rho = 0$*
- 2) *Pulse π with $\rho = \pi + \alpha/2$*



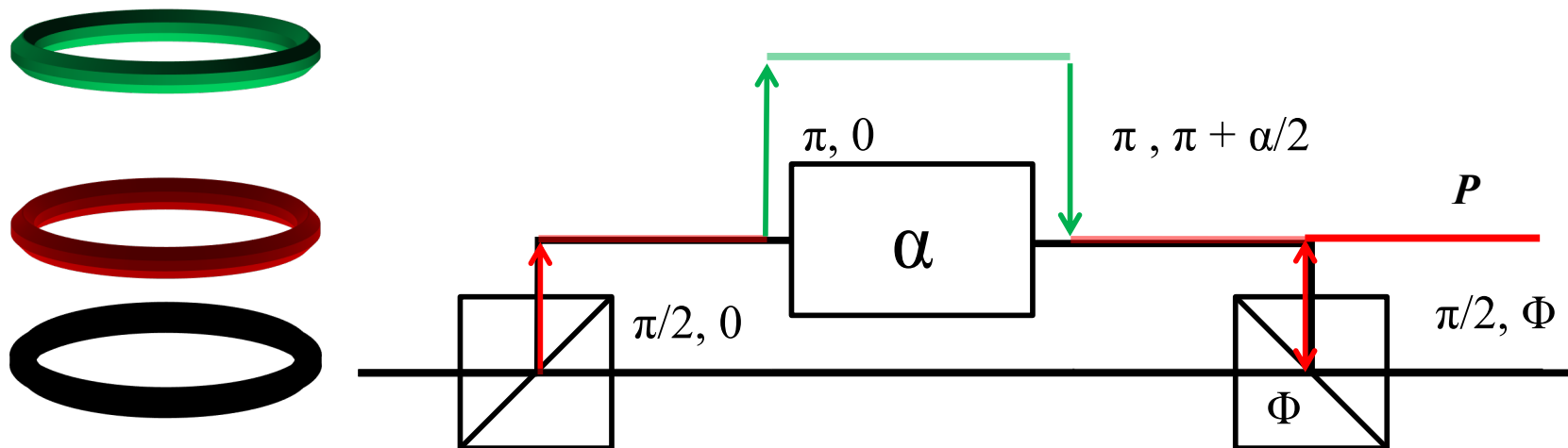
Geometric phase (Berry's phase)

Geometric phase accumulation :

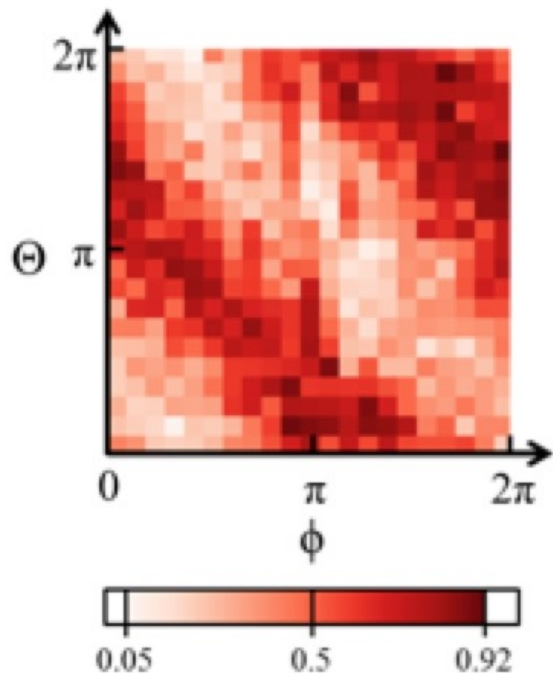
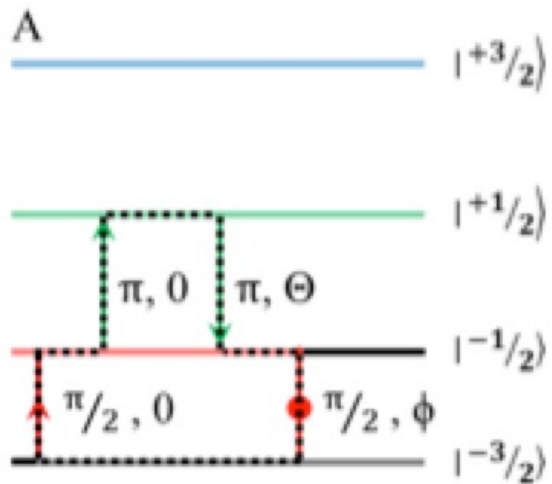
- 1) Pulse π with $\rho = 0$
- 2) Pulse π with $\rho = \pi + \alpha/2$



Phase measurement : Interferometer

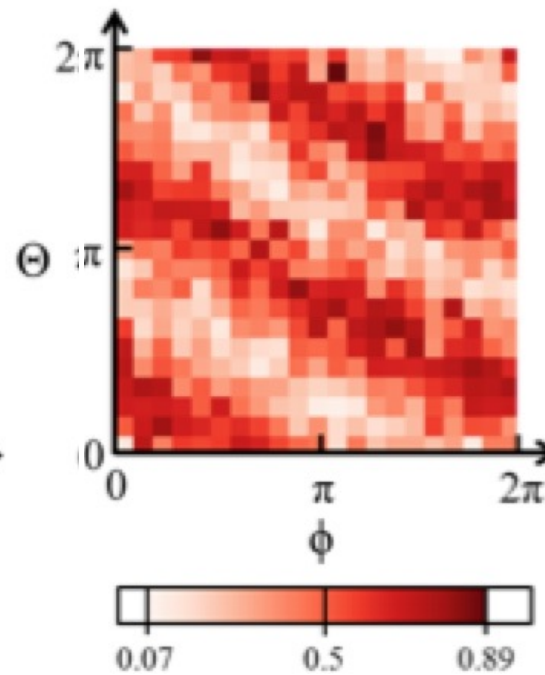
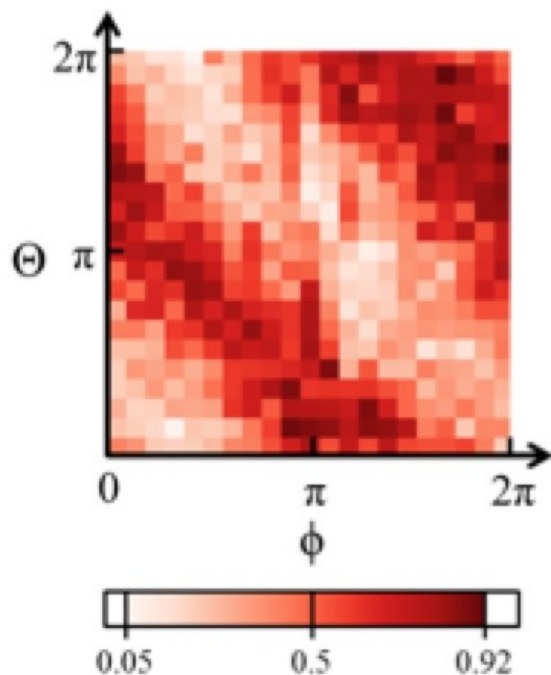
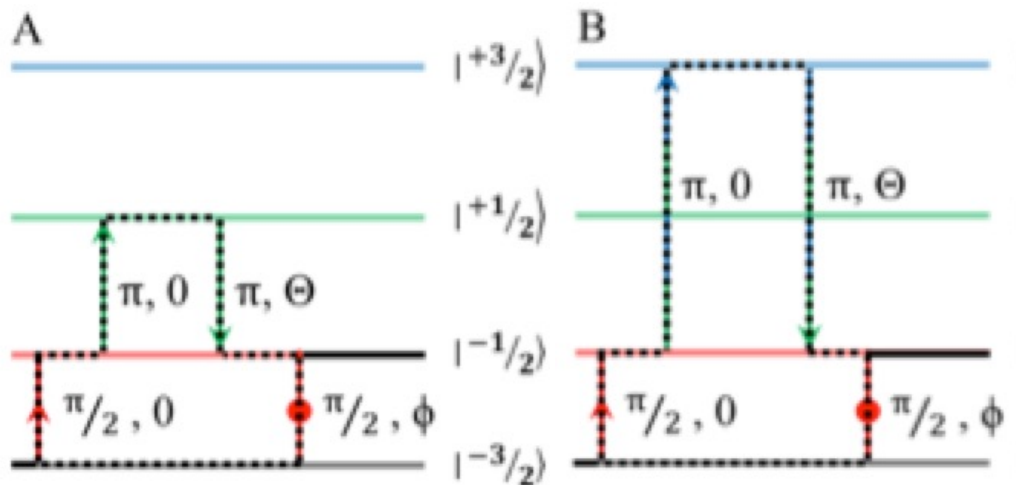


Geometric phase (Berry's phase)



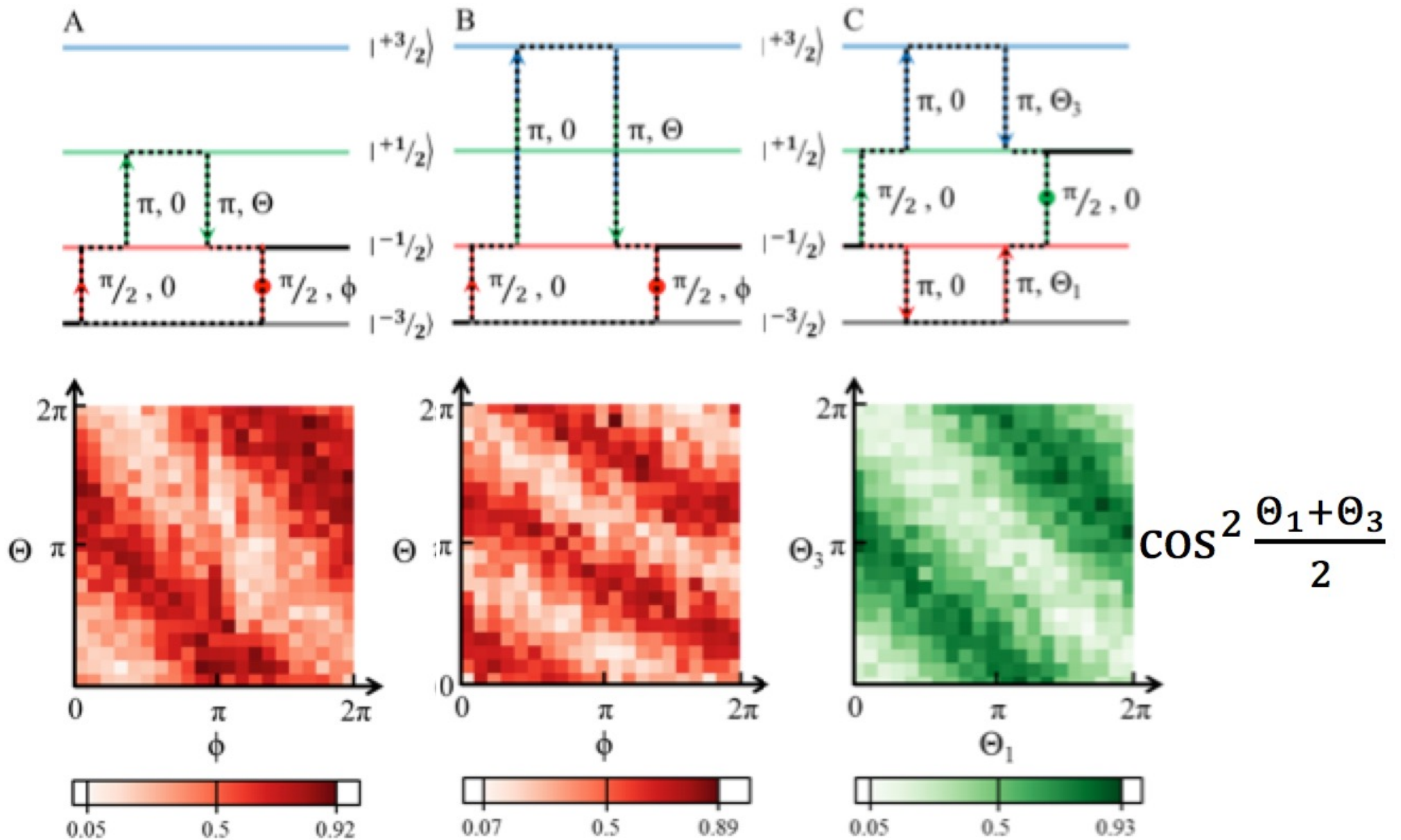
$$\cos^2 \frac{\Theta + \phi}{2} \cdot \text{interference}$$

Geometric phase (Berry's phase)



$$\cos^2 \frac{2\Theta + \phi}{2} \text{ interference}$$

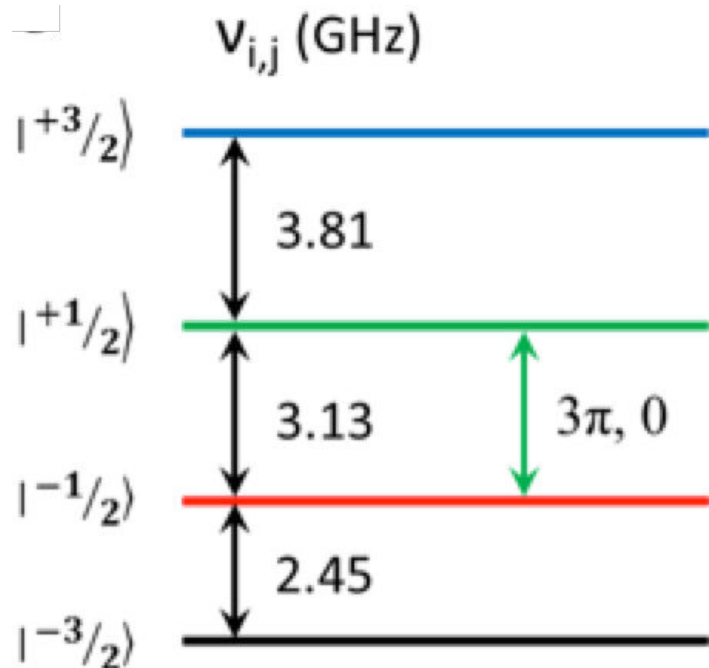
Geometric phase (Berry's phase)



iSWAP quantum gate

$$U_{iSWAP} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & i & 0 \\ 0 & i & 0 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}$$

$$= \begin{bmatrix} 0 & i \\ i & 0 \end{bmatrix}^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)} \oplus \mathbb{1}^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)} = R_x^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)}(3\pi)$$

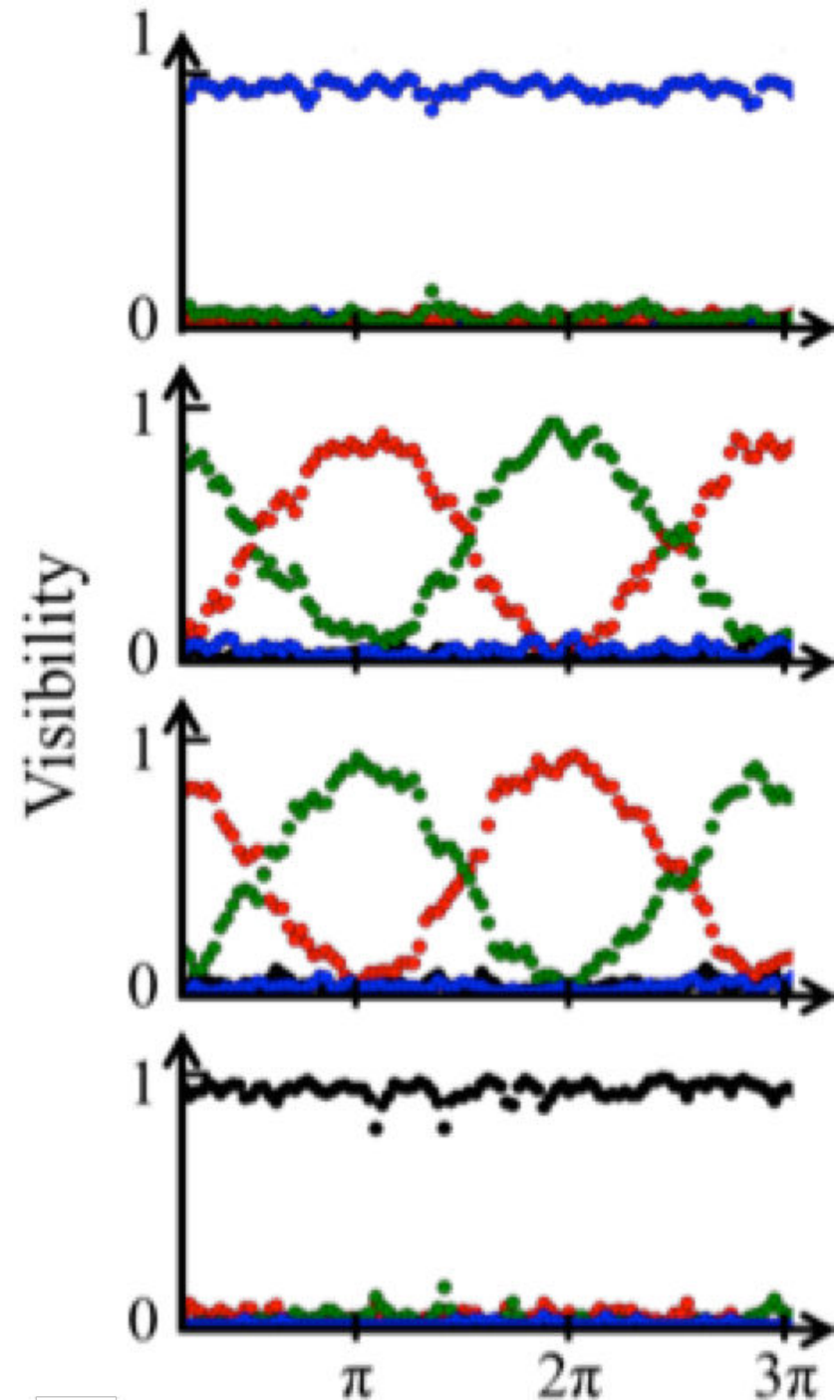
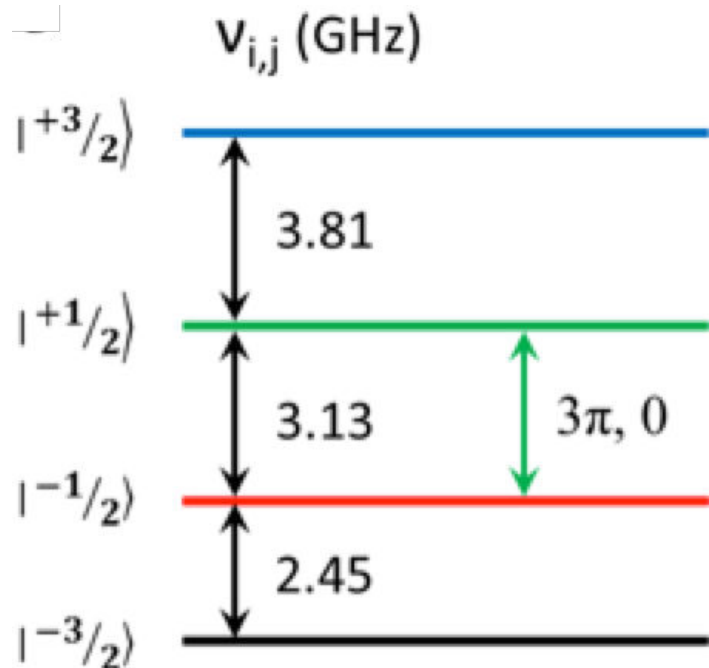


$$\begin{aligned} |3/2\rangle &\leftrightarrow |1\rangle_A \otimes |1\rangle_B \\ |1/2\rangle &\leftrightarrow |1\rangle_A \otimes |0\rangle_B \\ |-1/2\rangle &\leftrightarrow |0\rangle_A \otimes |1\rangle_B \\ |-3/2\rangle &\leftrightarrow |0\rangle_A \otimes |0\rangle_B \end{aligned}$$

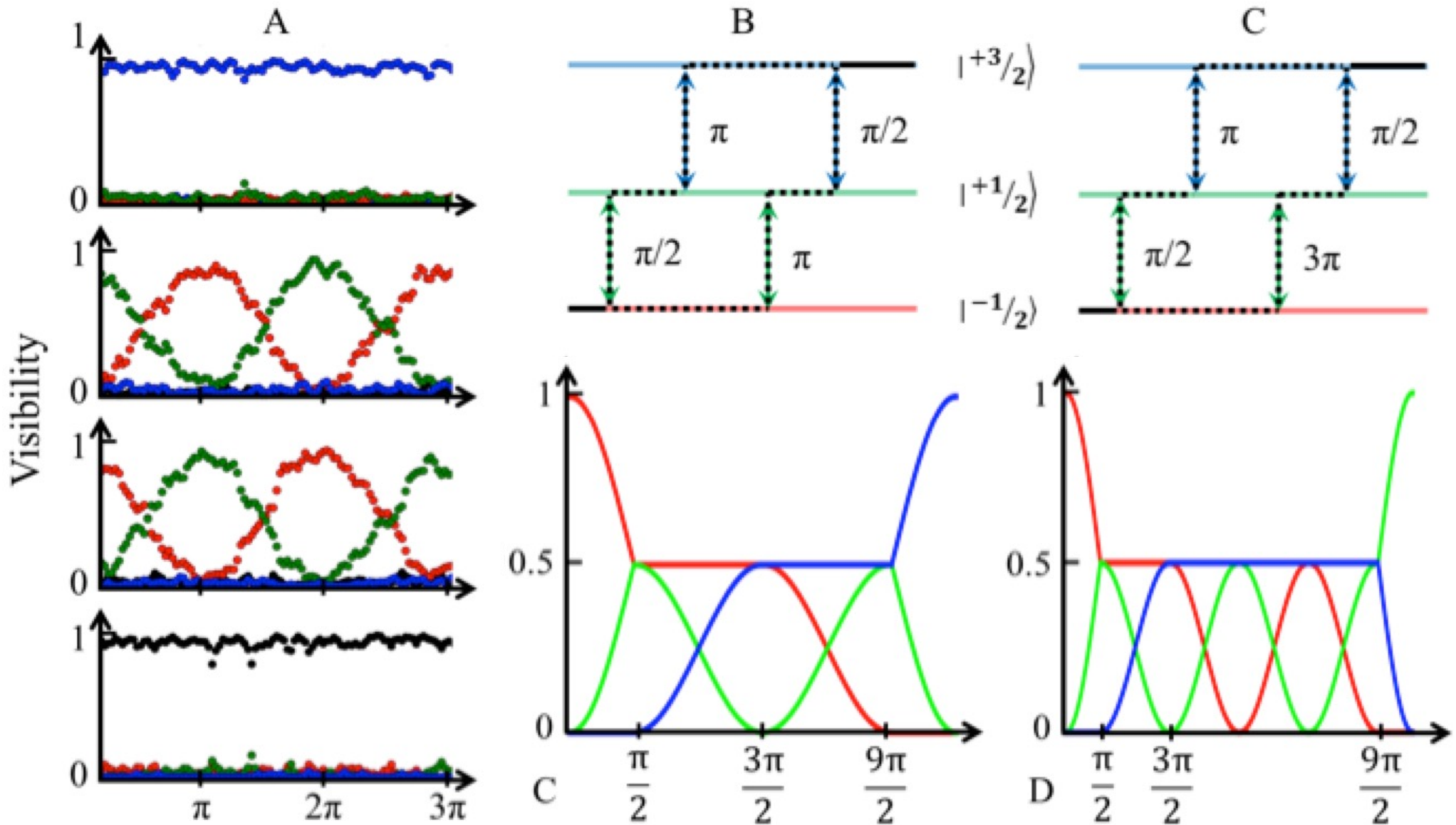
iSWAP quantum gate

$$U_{iSWAP} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & 0 & i & 0 \\ 0 & i & 0 & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix}$$

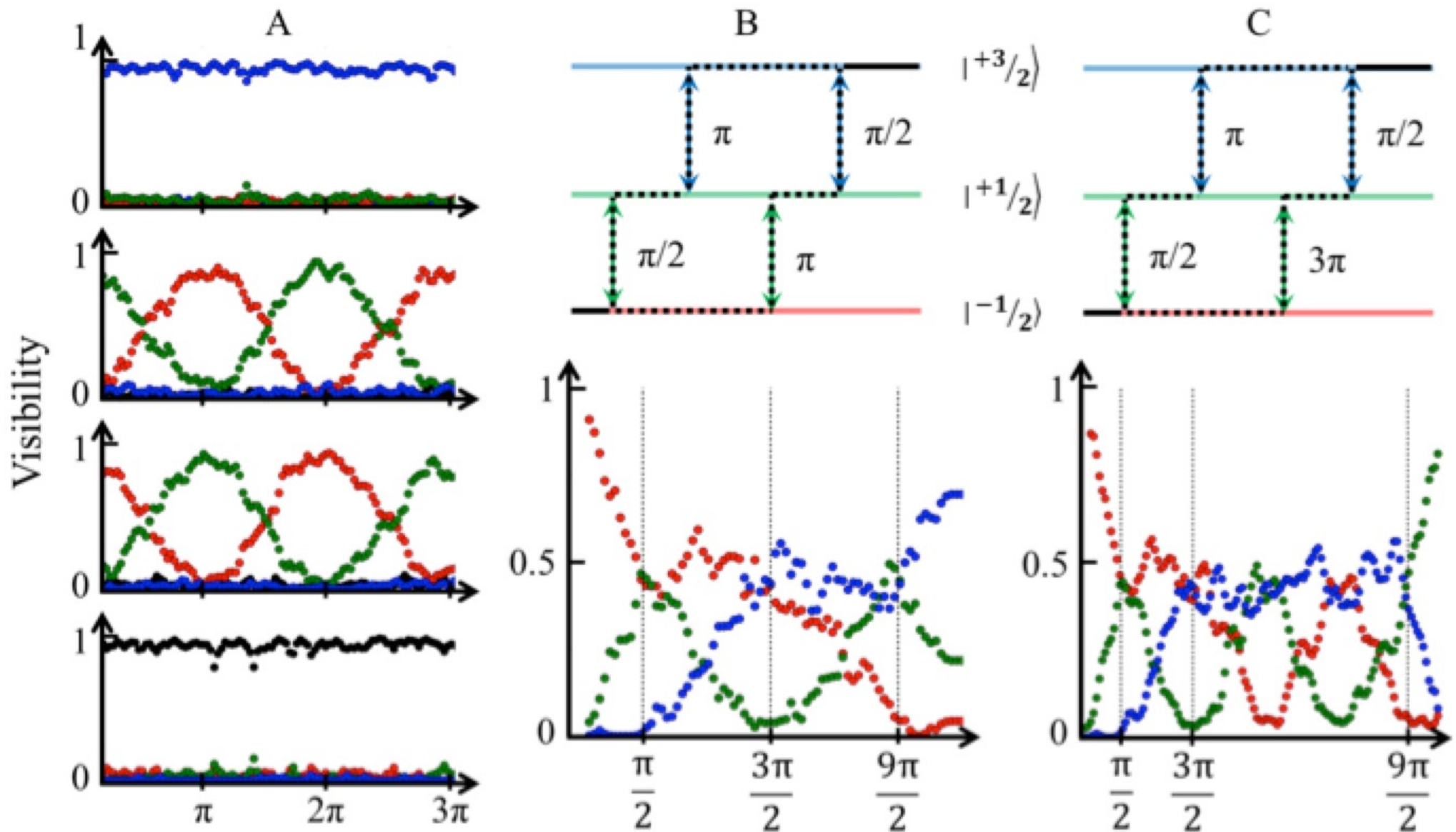
$$= \begin{bmatrix} 0 & i \\ i & 0 \end{bmatrix}^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)} \oplus \mathbb{1}^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)} = R_x^{(|-\frac{1}{2}\rangle, |\frac{1}{2}\rangle)}(3\pi)$$



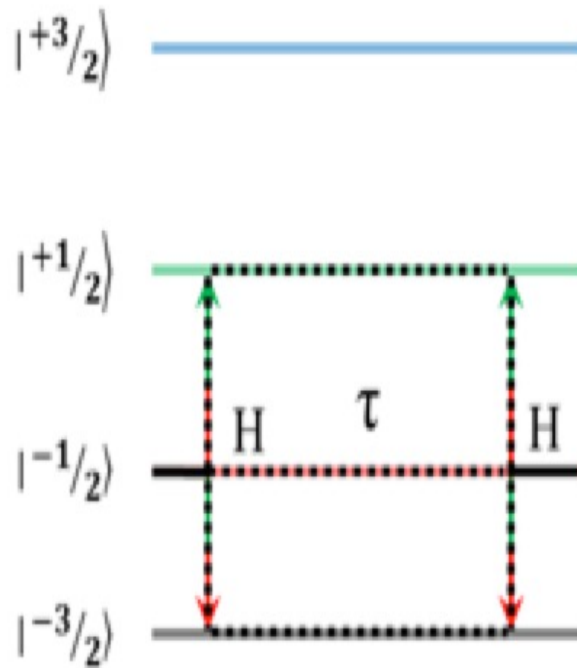
iSWAP quantum gate



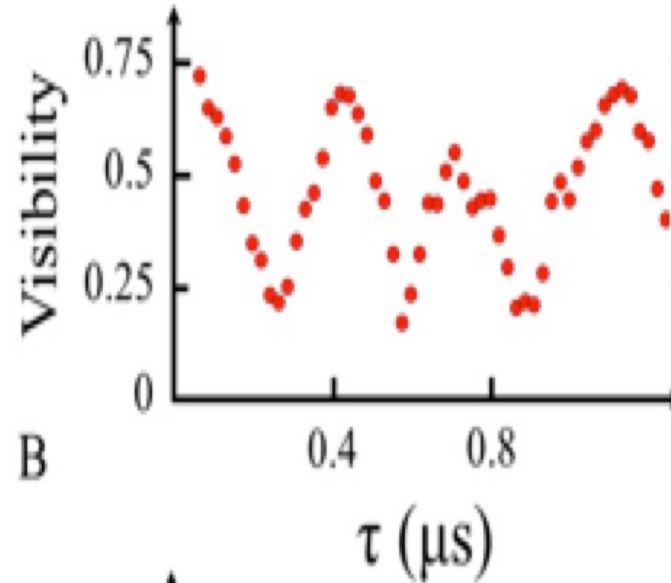
iSWAP quantum gate



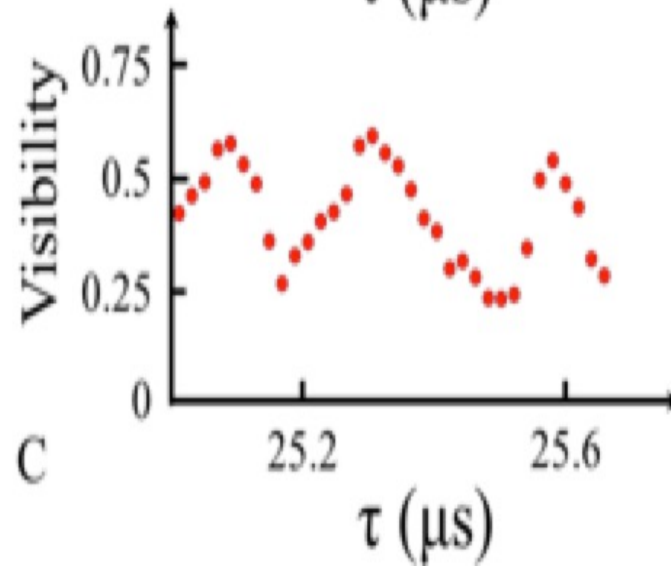
3-state coherence time



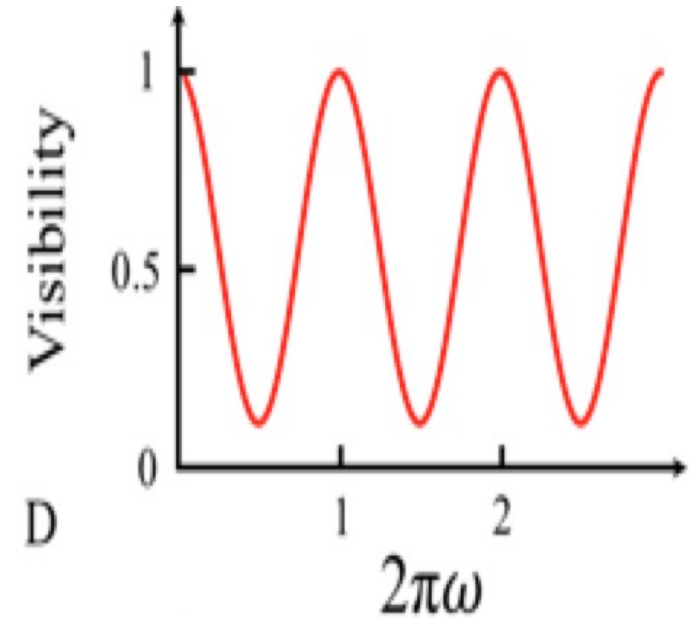
A



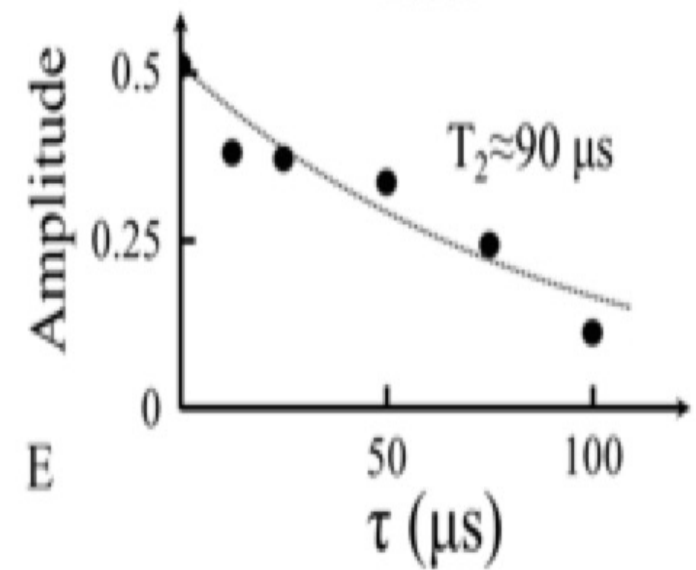
B



C



D



E

Molecular spin quantum technology?

Quantum computing in molecular magnets

M.N. Leuenberger & D. Loss

Nature 410, 789 (2001)

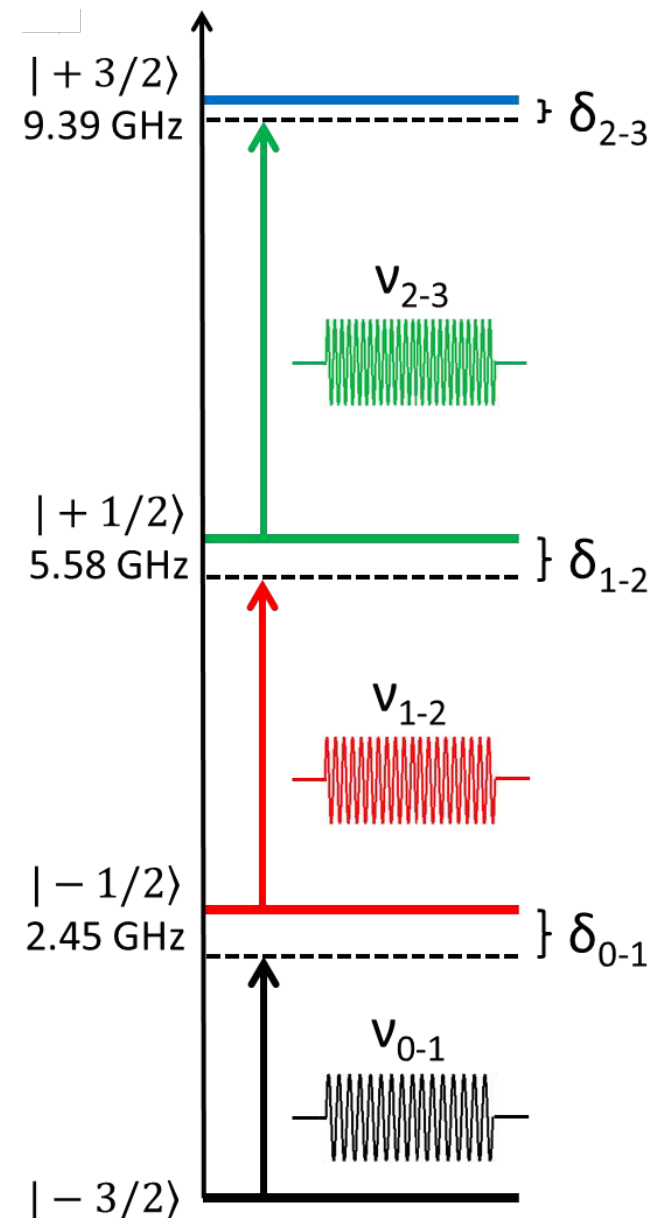
Grover algorithm for large nuclear spins in
semiconductors

M.N. Leuenberger & D. Loss

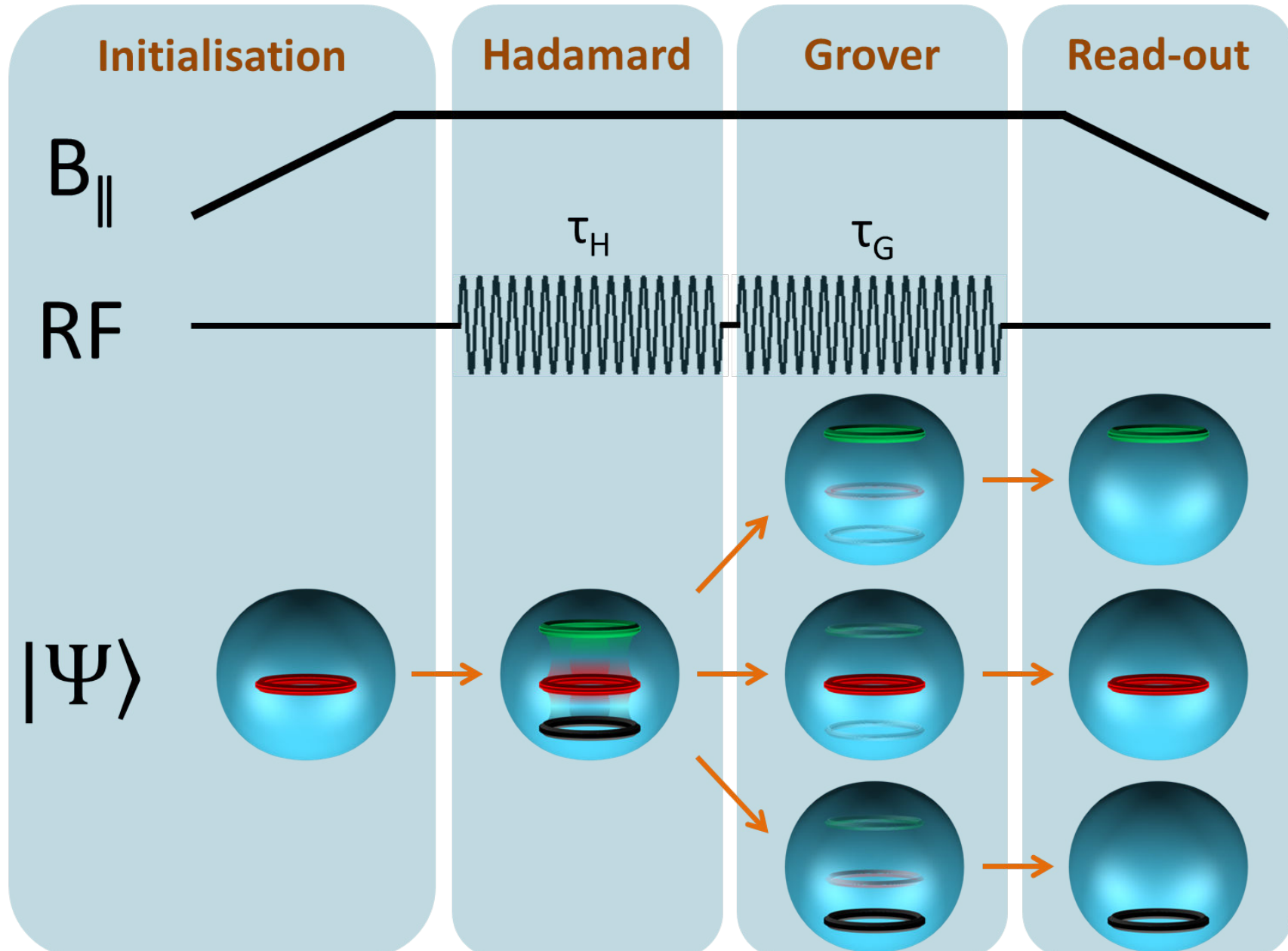
Physical Review B 68, 165317 (2003).



Grover's algorithm



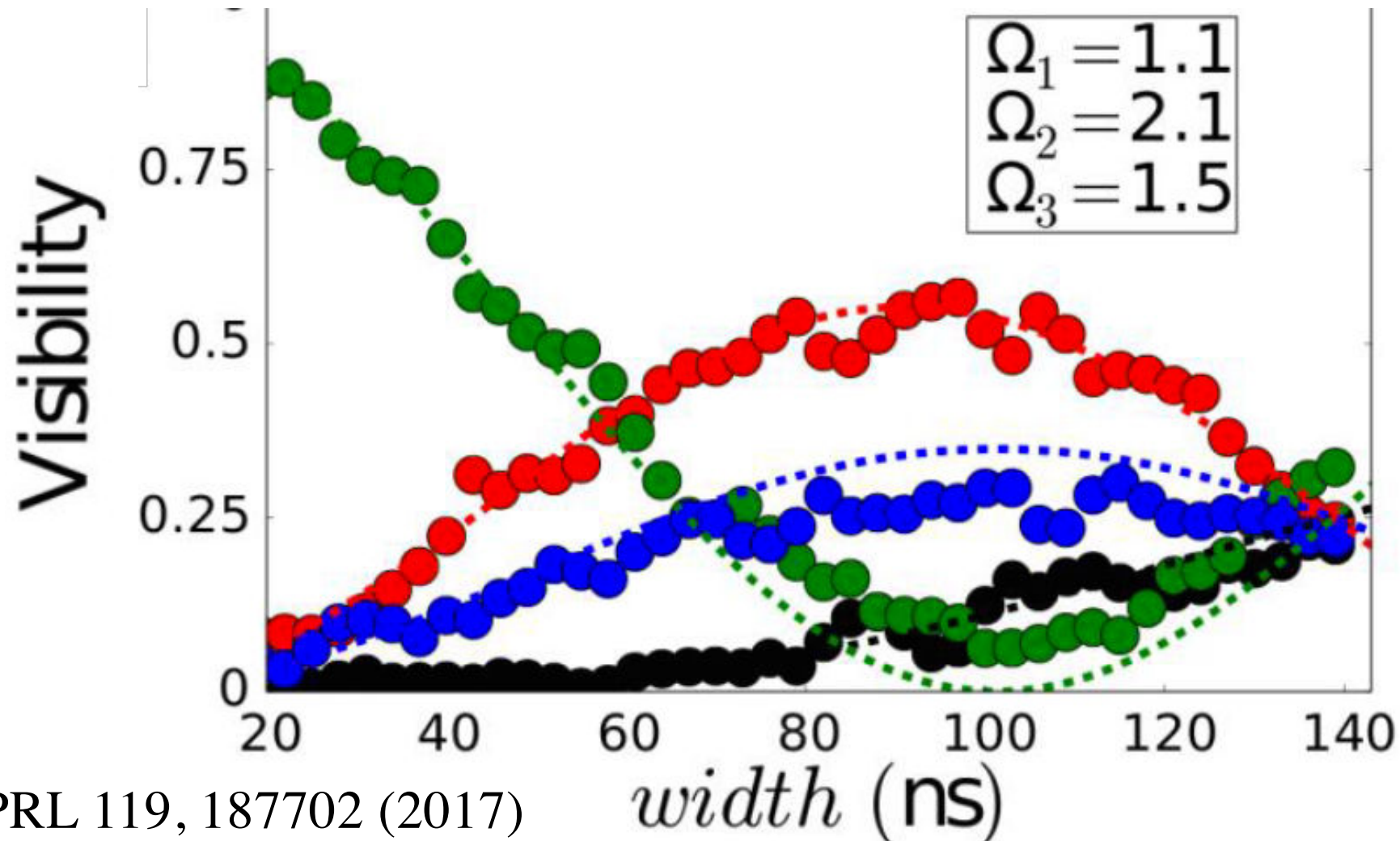
Grover's algorithm



Grover's algorithm

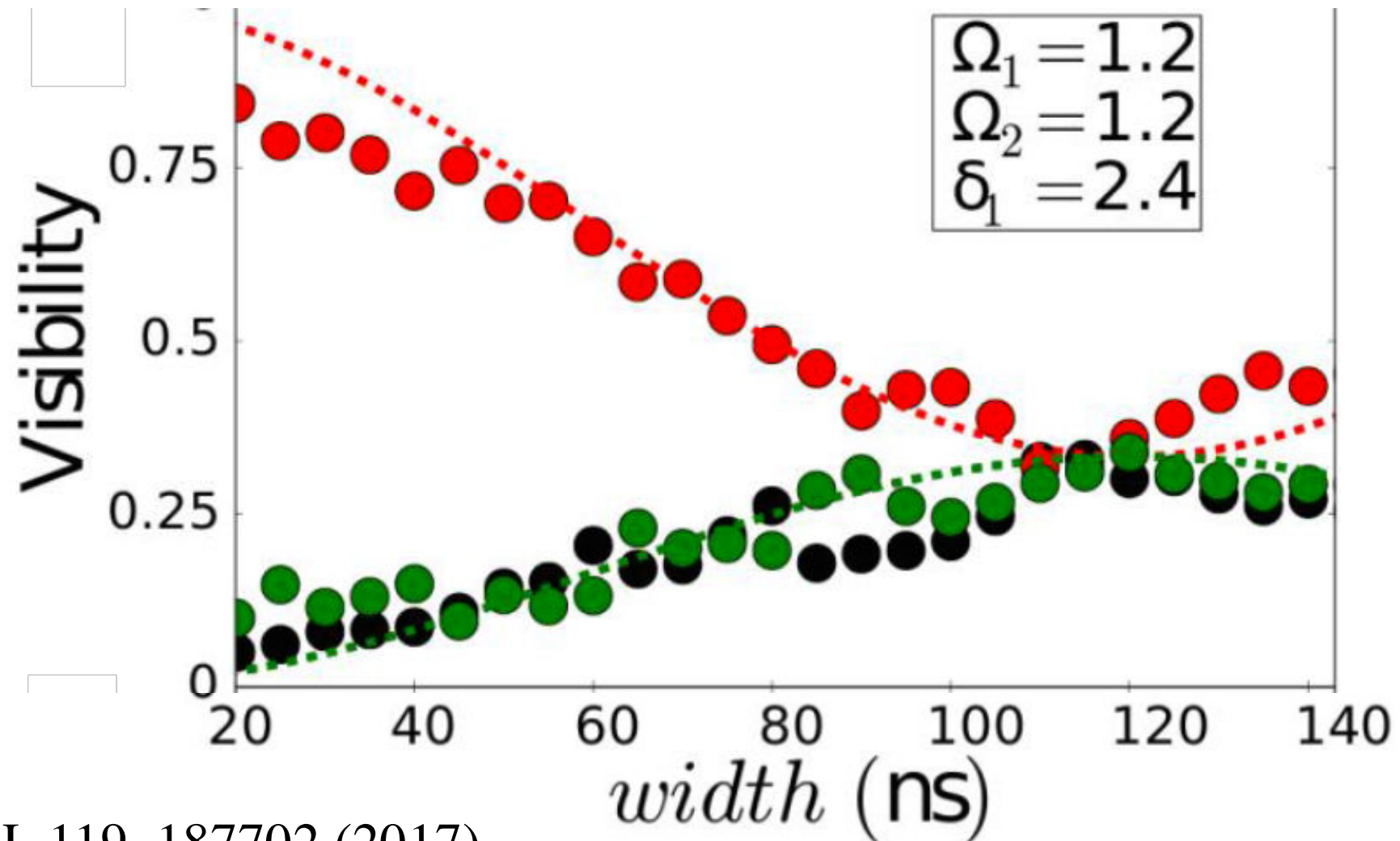
$$|+\frac{1}{2}\rangle \longrightarrow \frac{1}{\sqrt{4}}(|\frac{3}{2}\rangle + |\frac{1}{2}\rangle + |-\frac{1}{2}\rangle + |-\frac{3}{2}\rangle)$$

Hadamard gate with 4 states



Grover's algorithm

Hadamard gate with 3 states



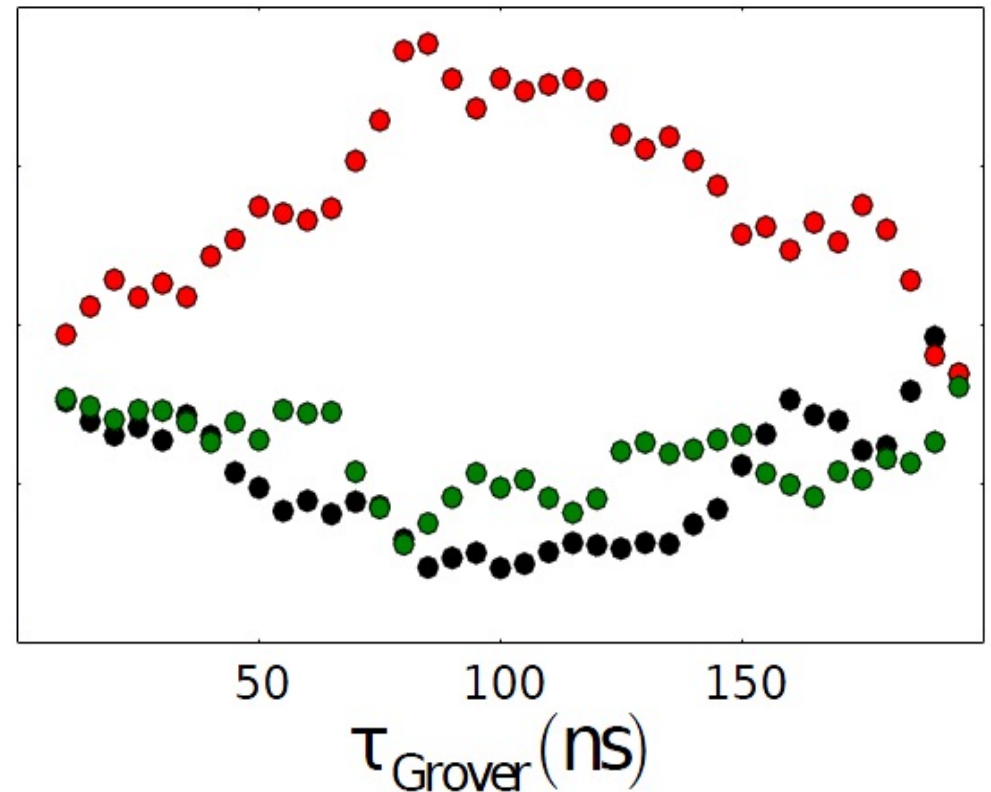
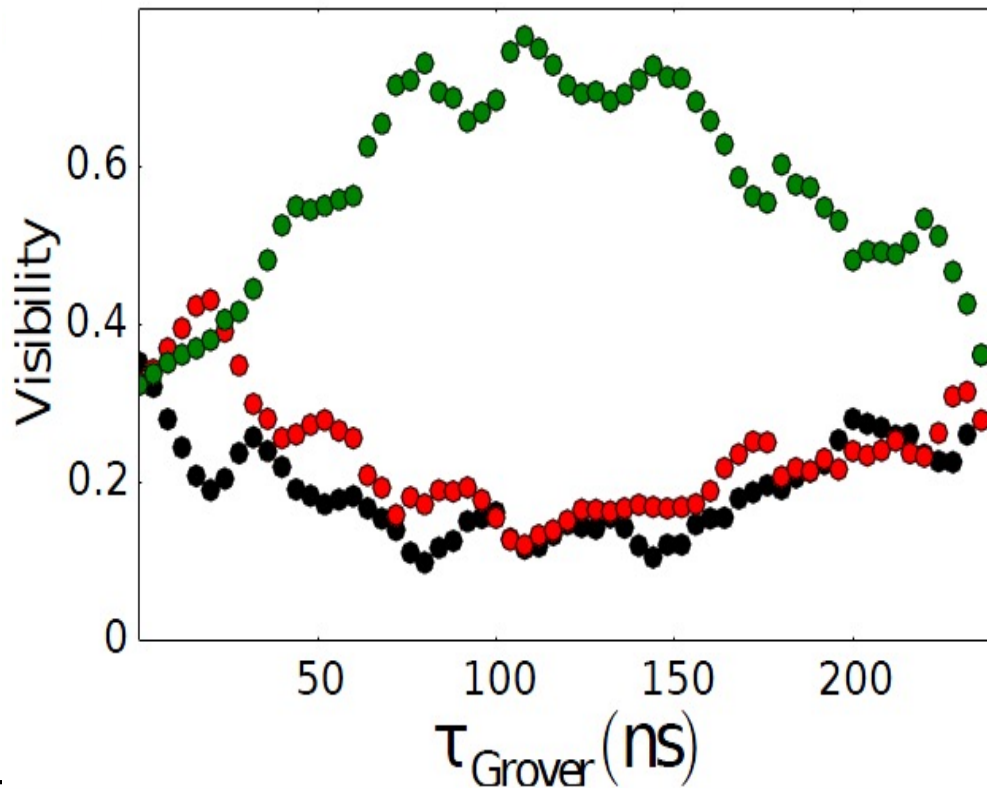
PRL 119, 187702 (2017)

Grover's algorithm

Selecting an element (state)

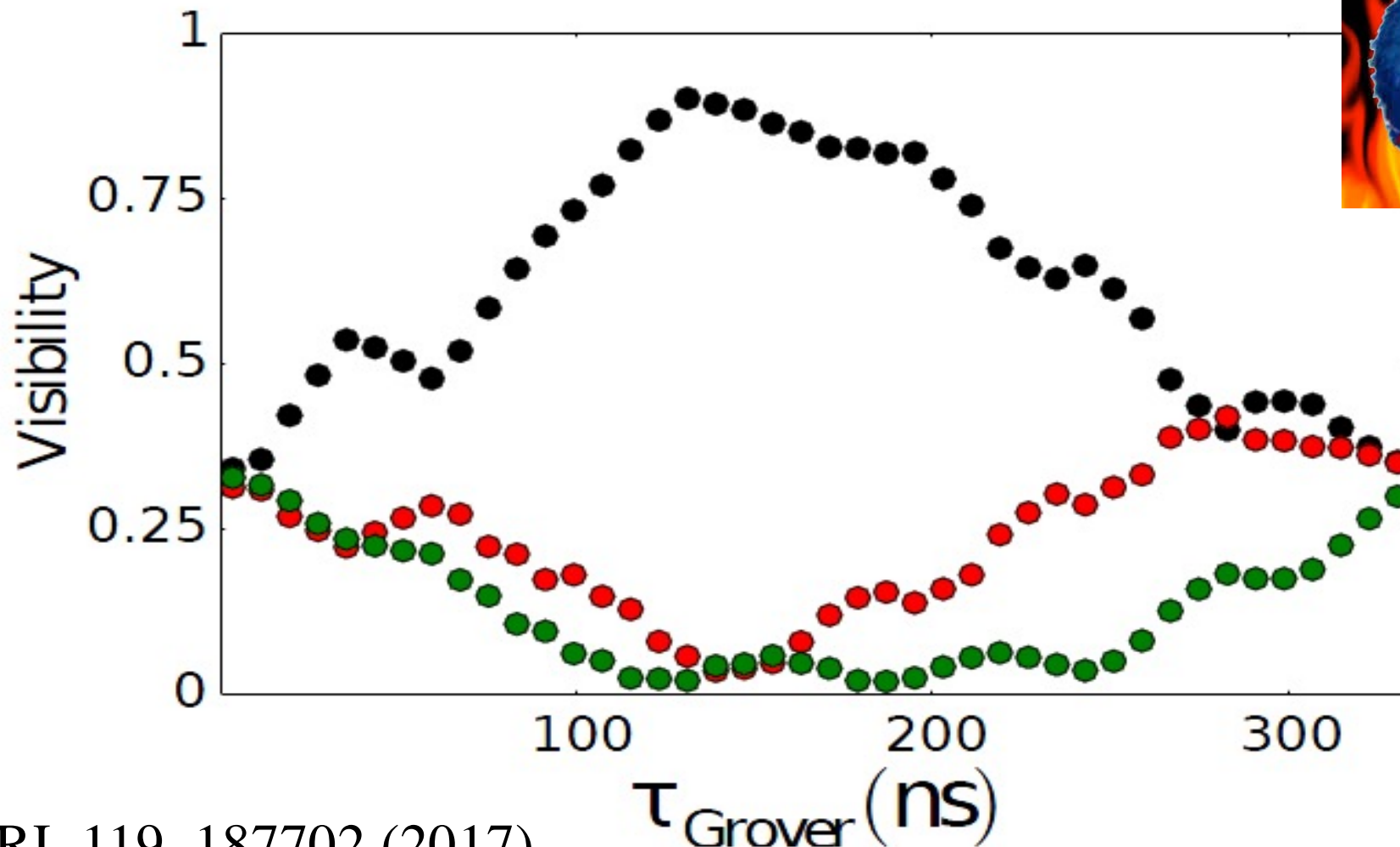


PRL 119, 187702 (2017)



Grover algorithm

Selecting an element (state)

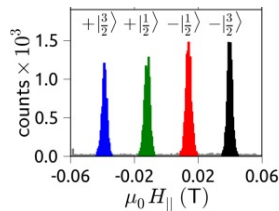


PRL 119, 187702 (2017)

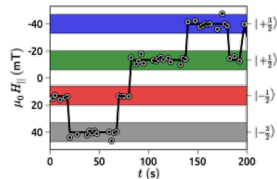
Single-molecule magnet qubits



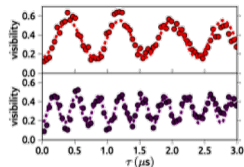
✓ **isolation:** intrinsic to nuclear spins



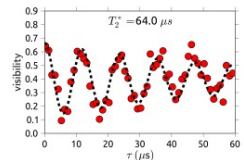
✓ **read-out:** through the position of the conductance jumps



✓ **initial state preparation:** by measurement due to the QND nature of the detection



✓ **coherent manipulation:** we could perform simple quantum operations

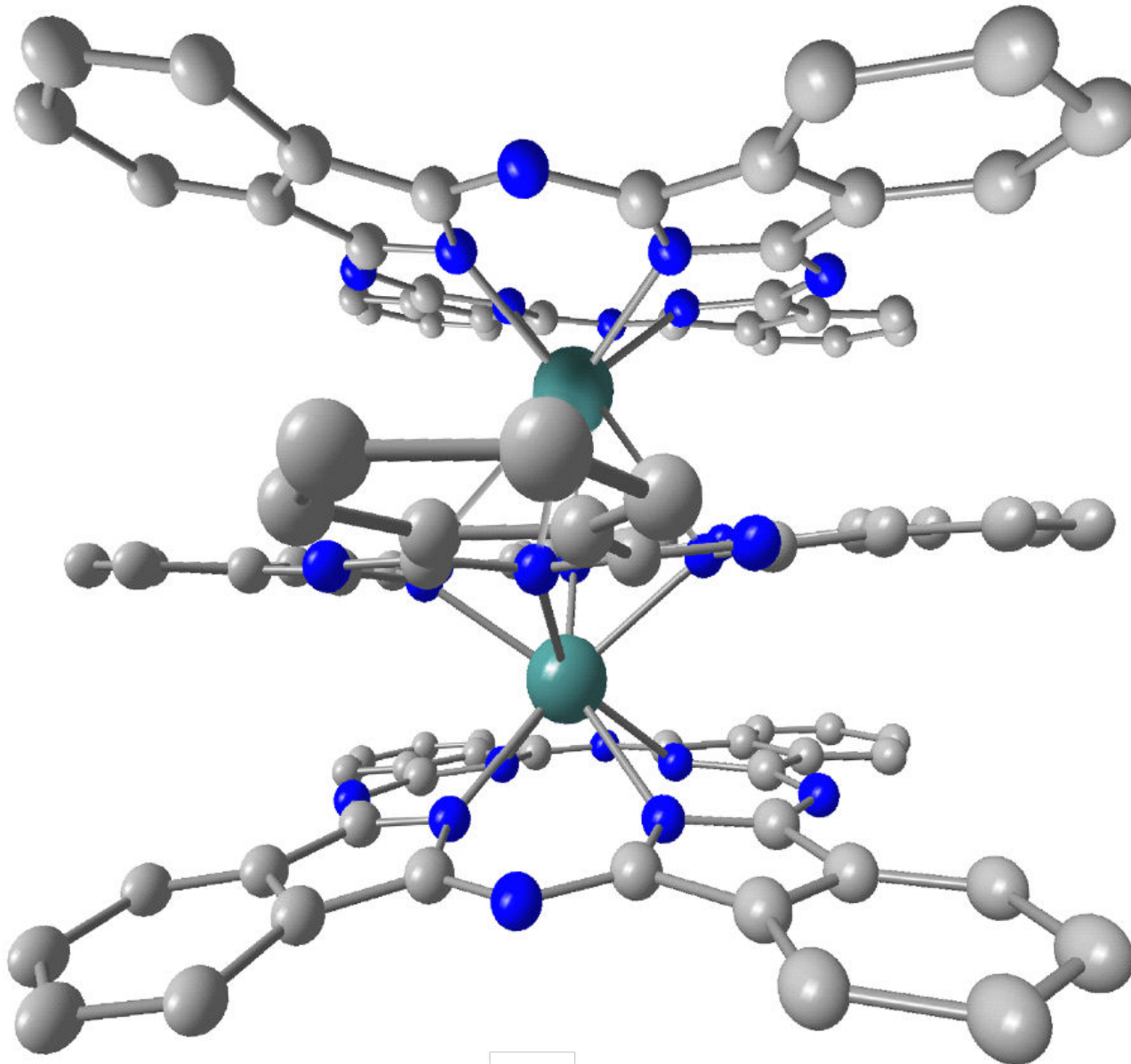


✓ **information storage:** quantum information was stored for $>300 \mu s$

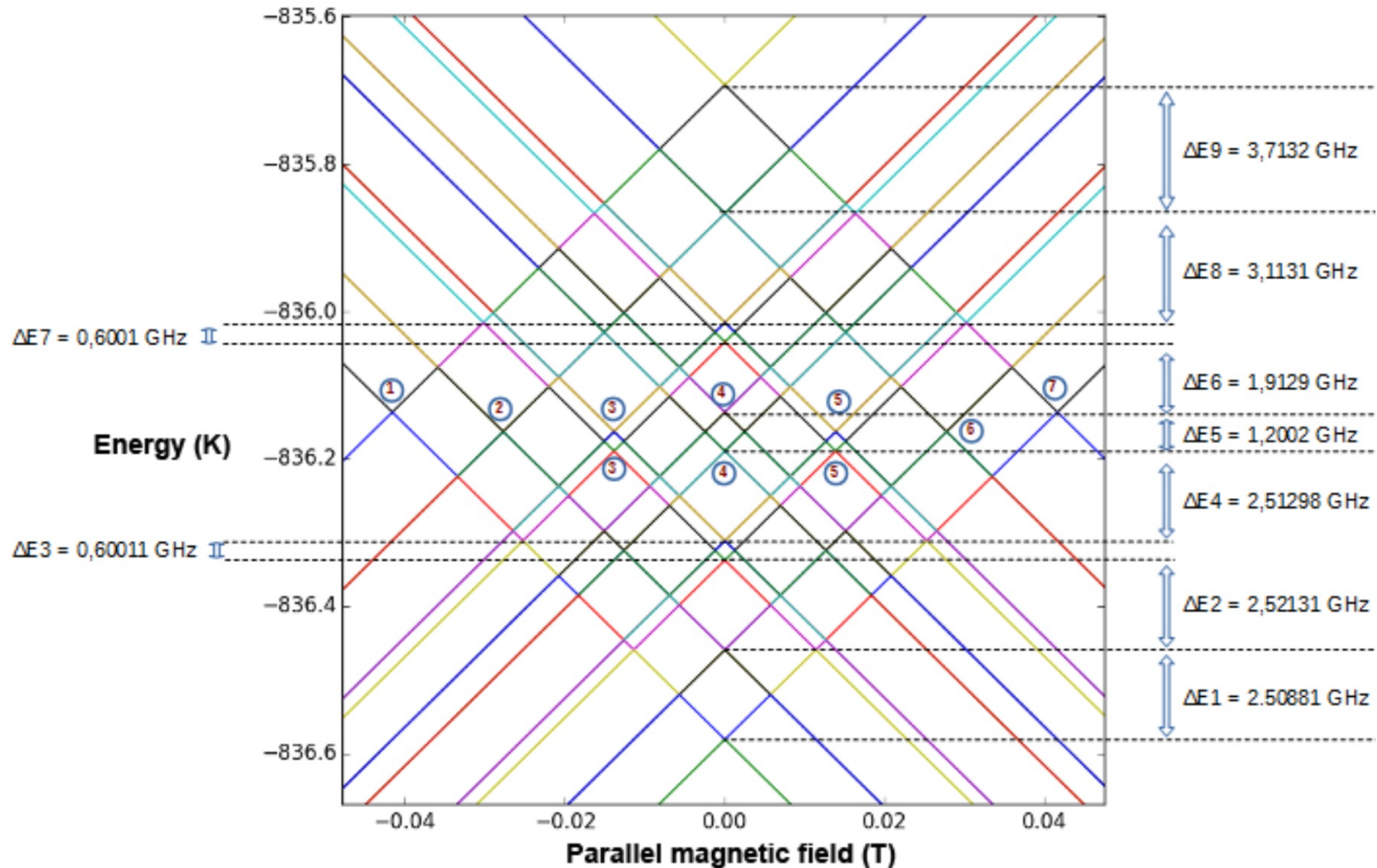


rover's algorithm, quantum gates

Triple Decker Tb_2Pc_3



Triple Decker Tb_2Pc_3



Quantum computing in molecular magnets

Michael N. Leuenberger & Daniel Loss

NATURE, 410, 791 (2001)

- implementation of Grover's algorithm
- storage unit of a dynamic random access memory device.
- fast electron spin resonance pulses can be used to decode and read out stored numbers of up to 10^5 with access times as short as 0.1 nanoseconds.

