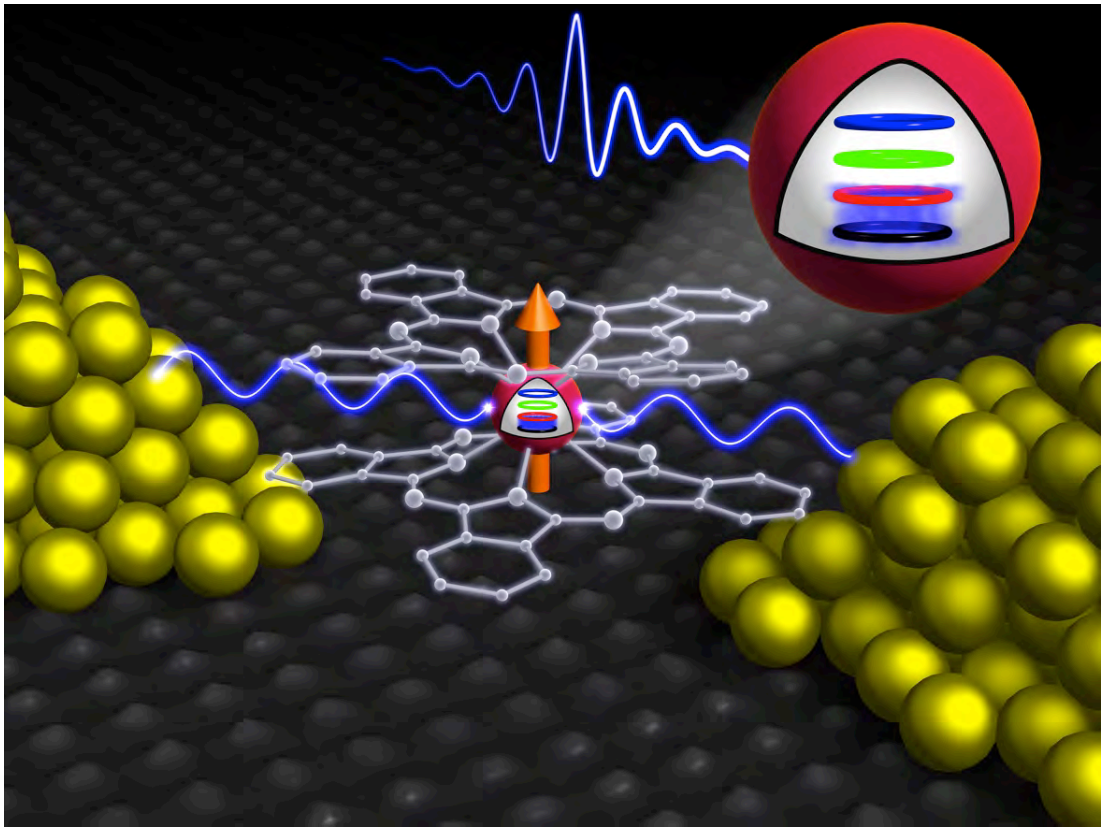
Reading out spin states of single molecules: application to Grover's algorithm

W. Wernsdorfer

Institut Néel, CNRS, Grenoble

Karlsruhe Institute of Technology (PHI, INT)



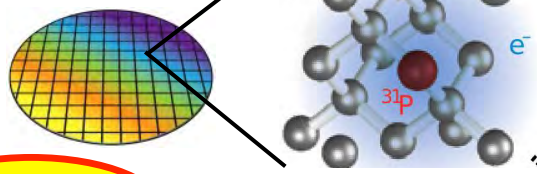
MoQuaS
Molecular Quantum Spintronics
FP7-ICT-2013-10

European initiative in quantum technologies

Quantum Technologies

Isolation
Initialization
Storage
Manipulation
Read-out

DiVincenzo 1996

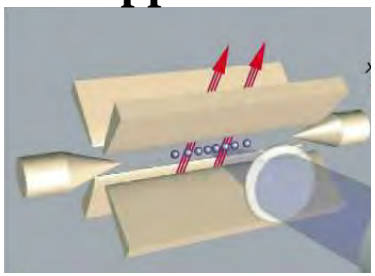


^{31}P in silicon

Intel

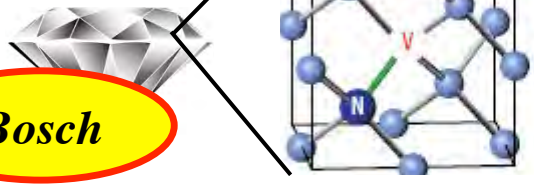
ture 489, 541 (2012)

trapped ions



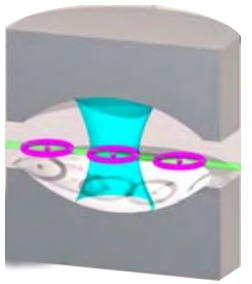
Nature 453, 1008 (2008)

NV center



Bosch

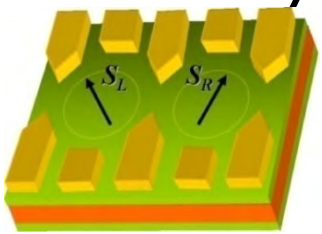
PRL 92, 076401 (2004)



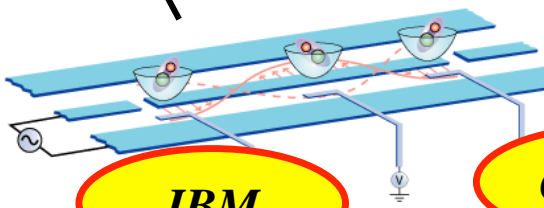
light in
a cavity

Nature 446, 275 (2007)

quantum
dots



Nature 491, 426 (2012) Nature 422, 766 (2006)



superconducting
circuits

IBM

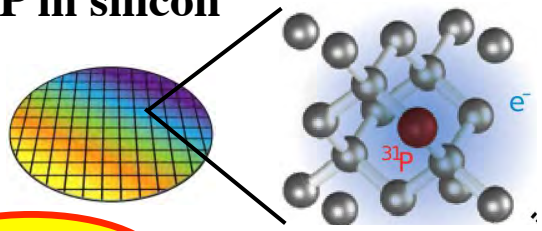
Google

D-wave

Nature 4

Quantum Technologies

^{31}P in silicon



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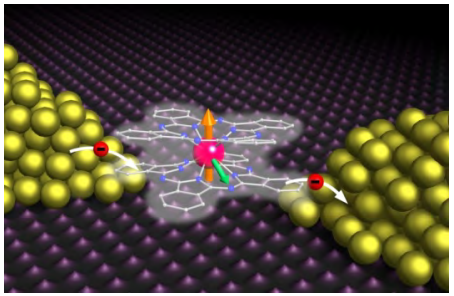
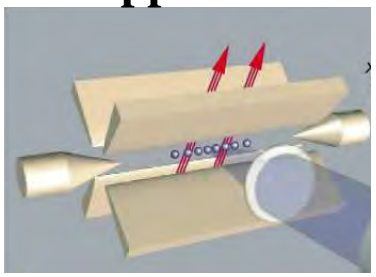


Nature 488, 357 (2012)
Science 344, 1135 (2014)

trapped ions



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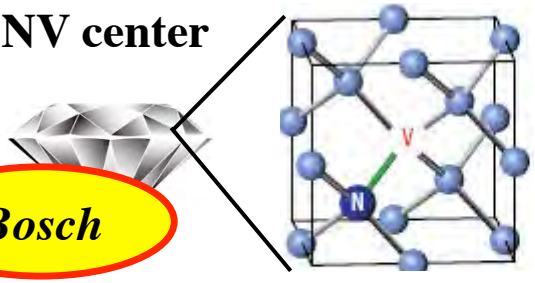

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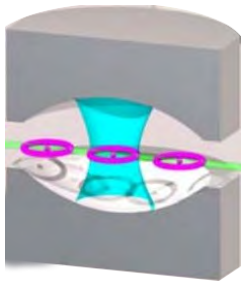

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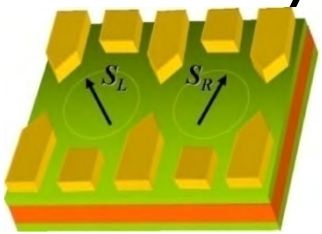

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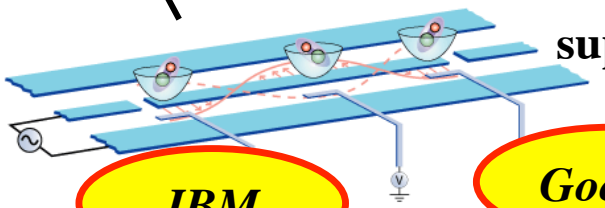

superconducting
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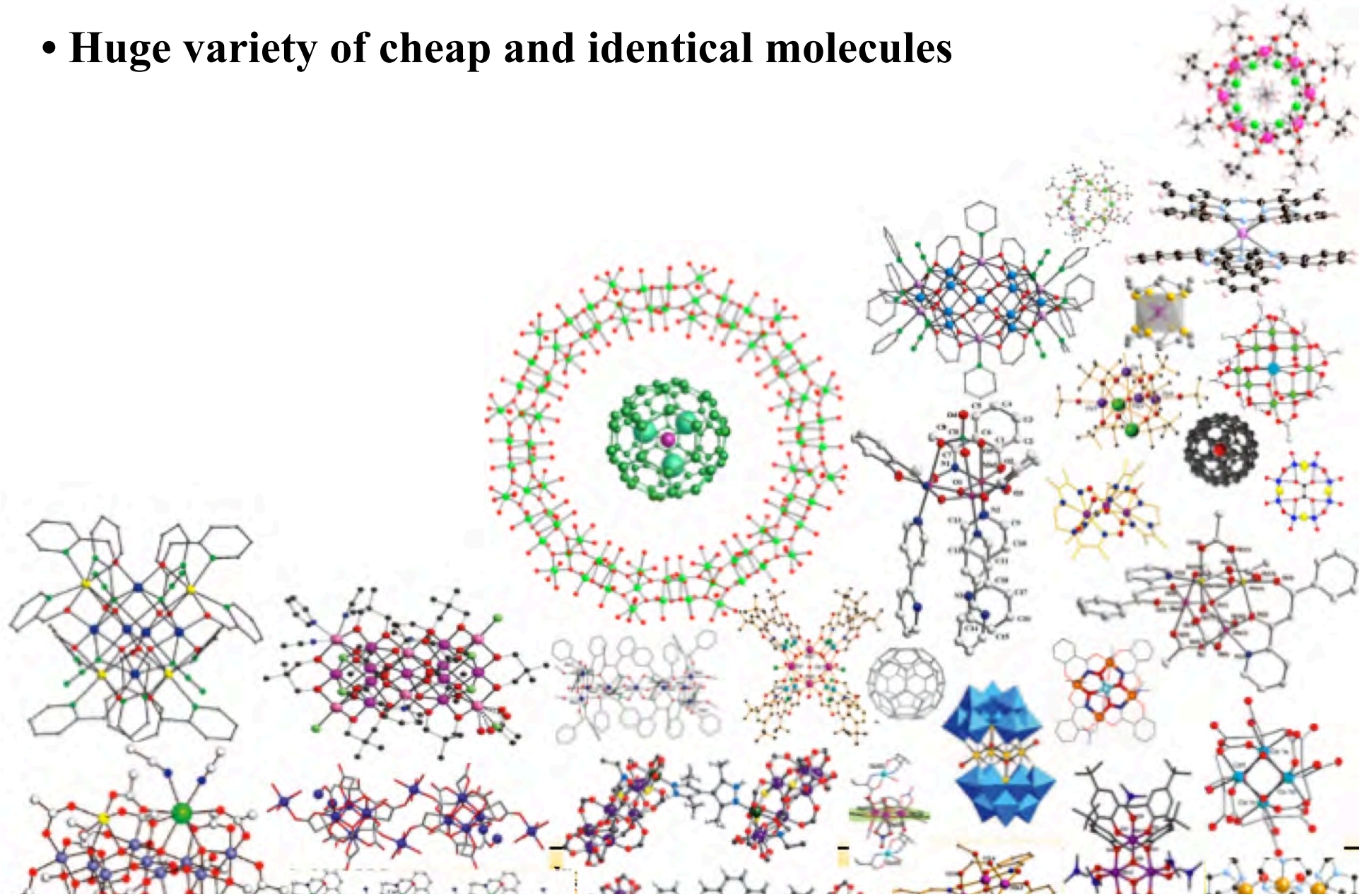
D-wave

Nature 4

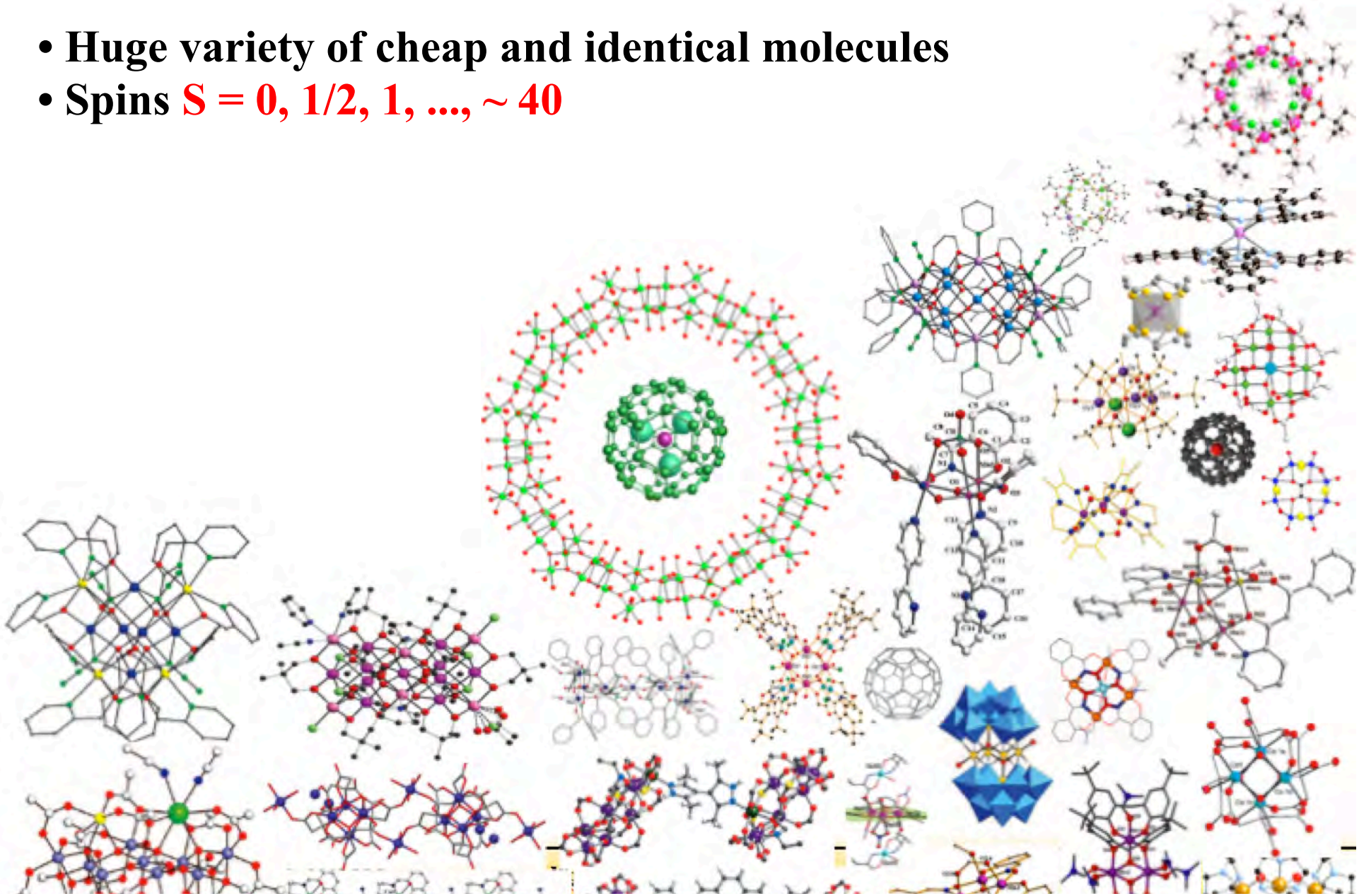
Molecular spin quantum technology?

- **Huge variety of cheap and identical molecules**



Molecular spin quantum technology?

- Huge variety of cheap and identical molecules
- Spins S = 0, 1/2, 1, ..., ~ 40



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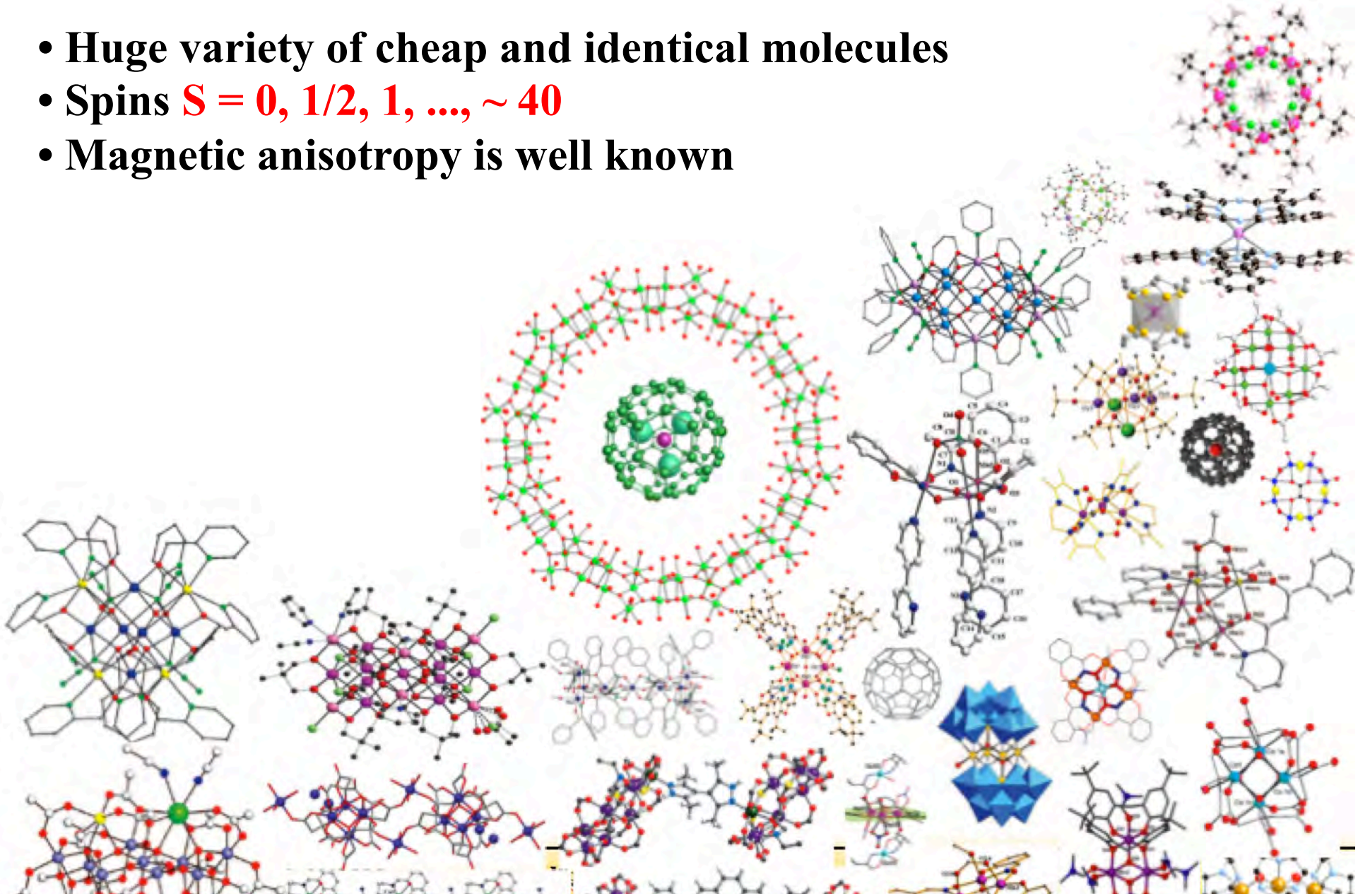

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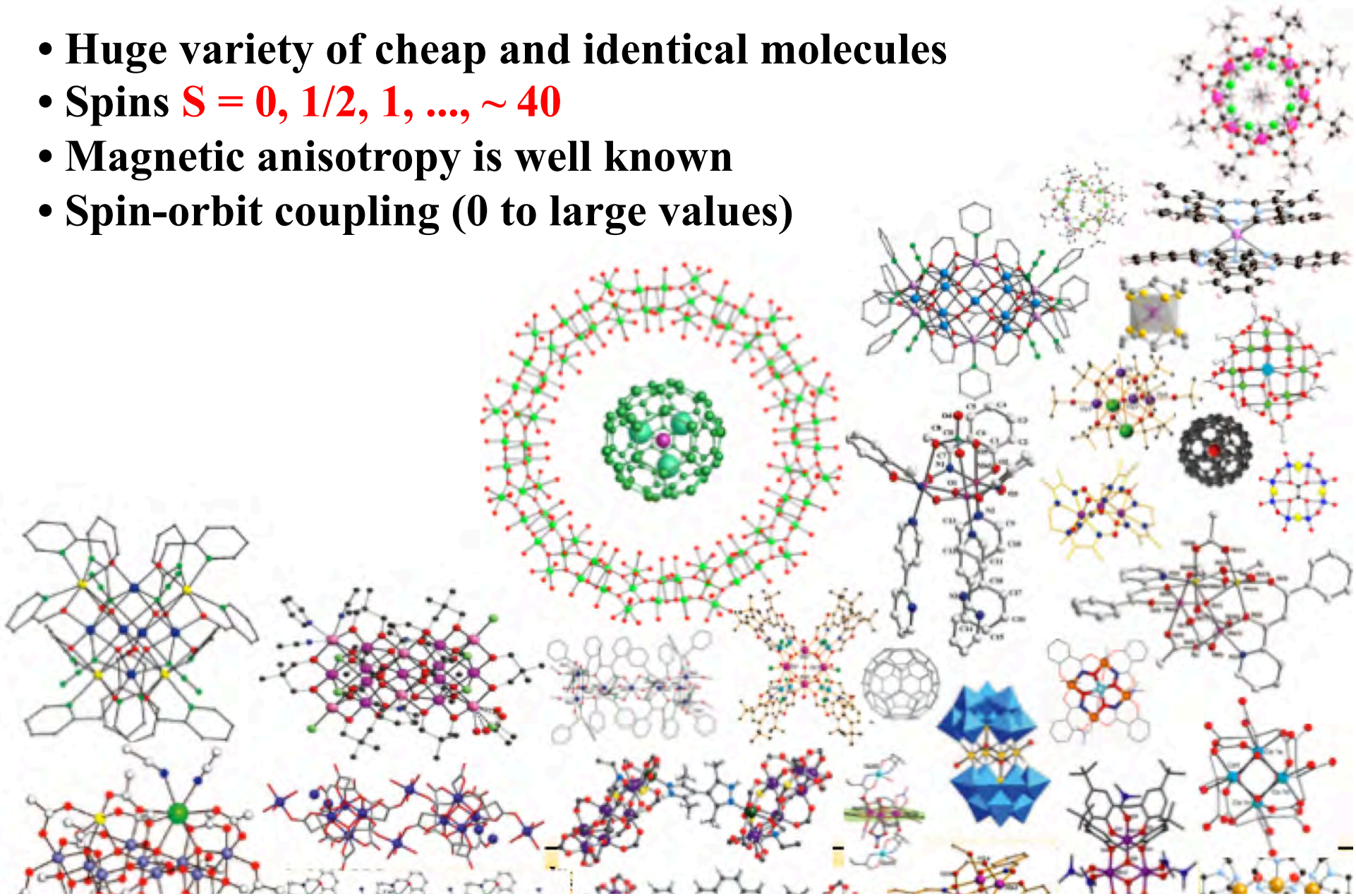

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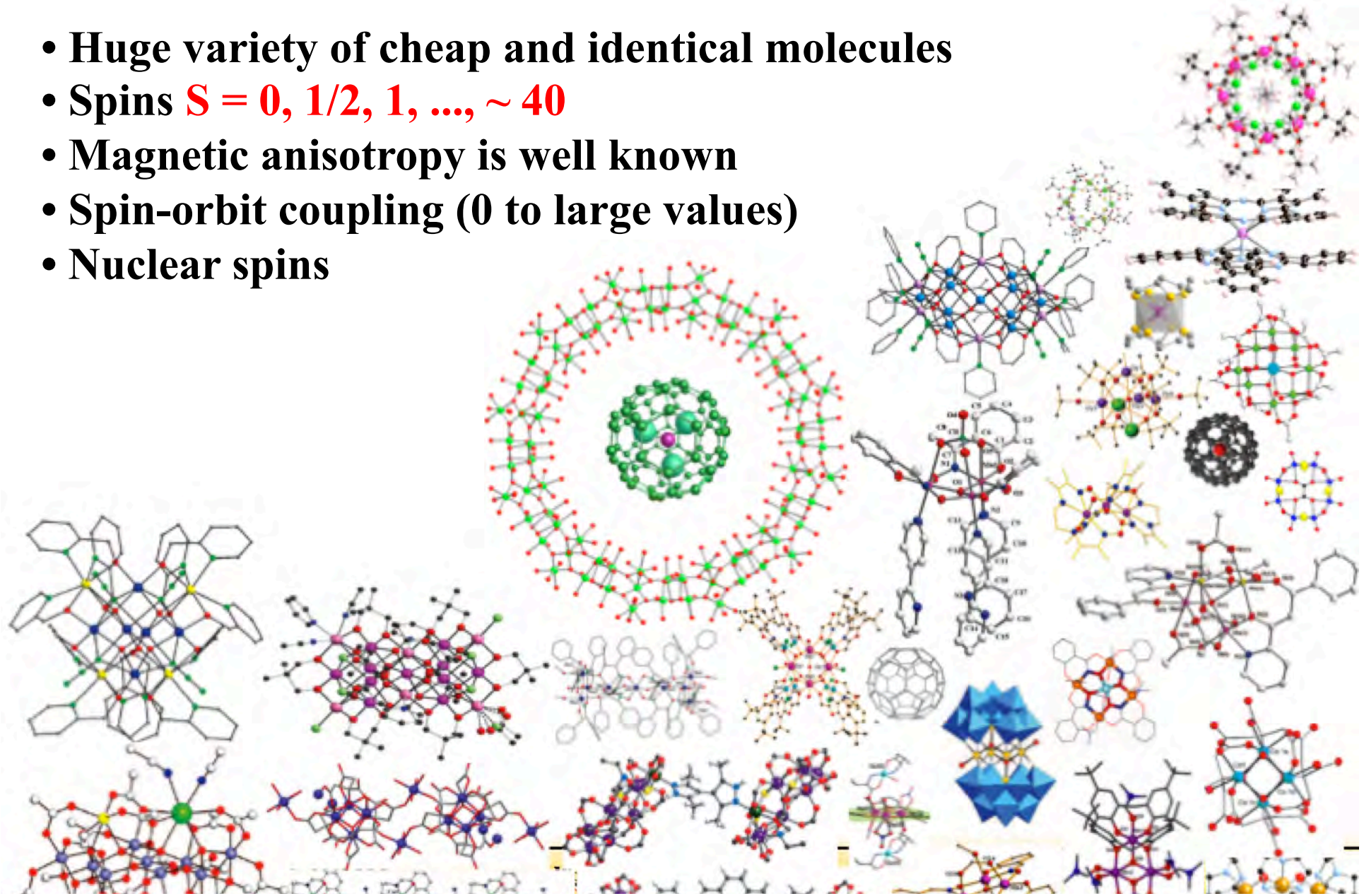

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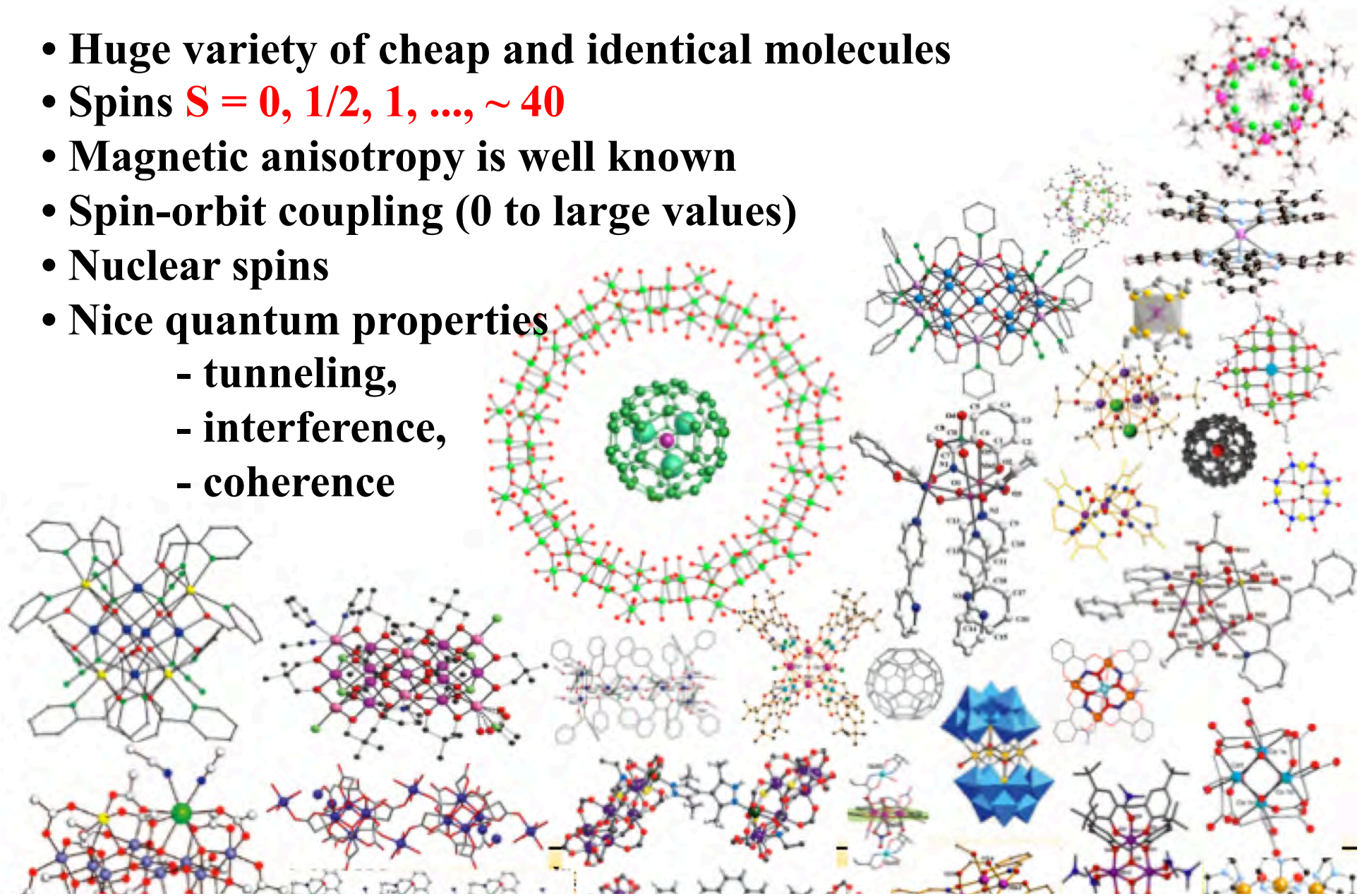
- Nice quantum properties
  - tunneling,
  - interference,
  - coherence



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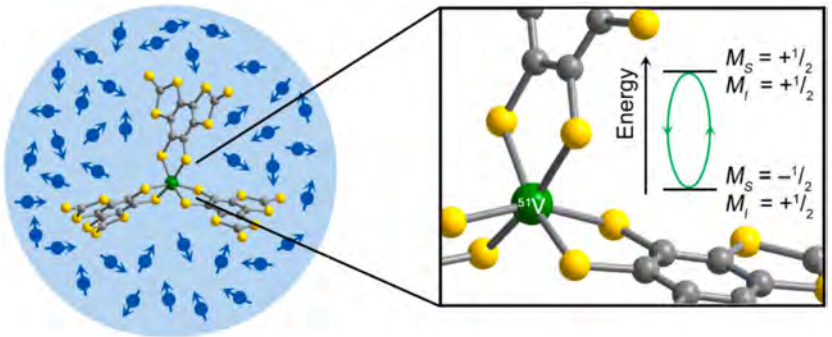
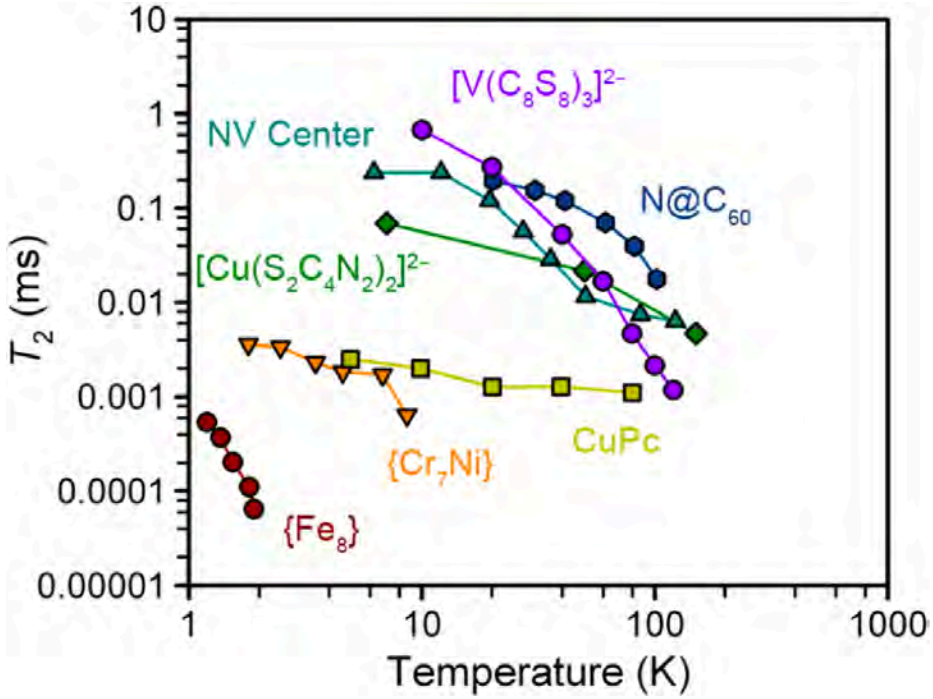
  T_1 up to years, T_2 ~ ms





ACS Cent. Sci. 1, 488 (2015)

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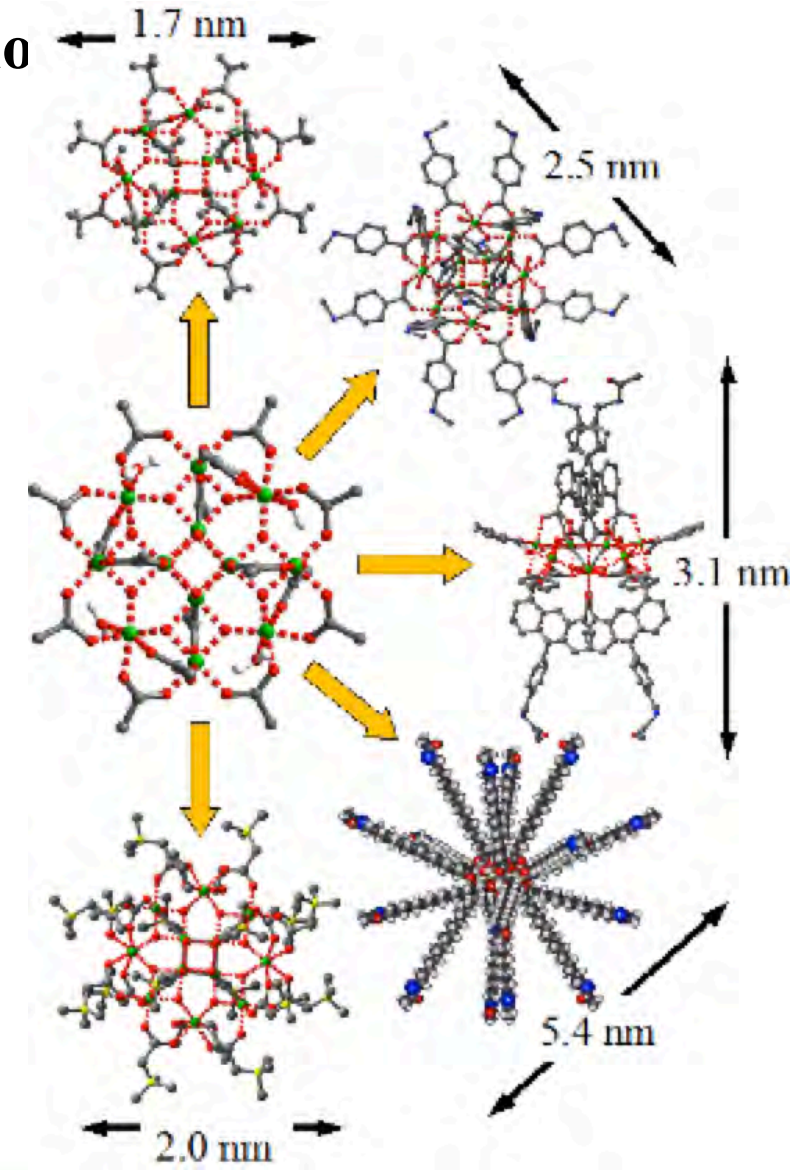
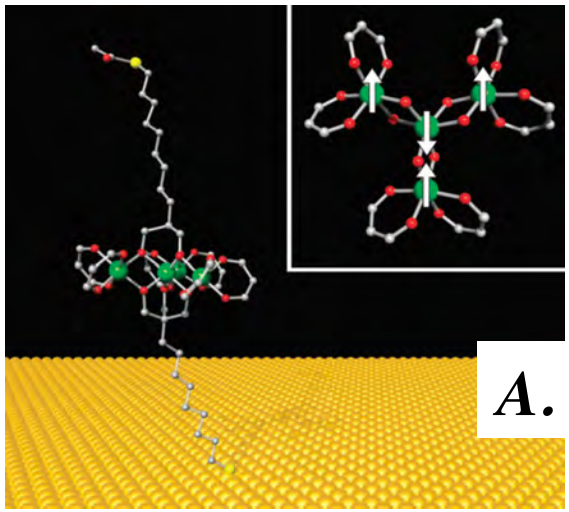
  T_1 up to years, T_2 ~ μs – ms
- Peripheral functionalization of SMMs

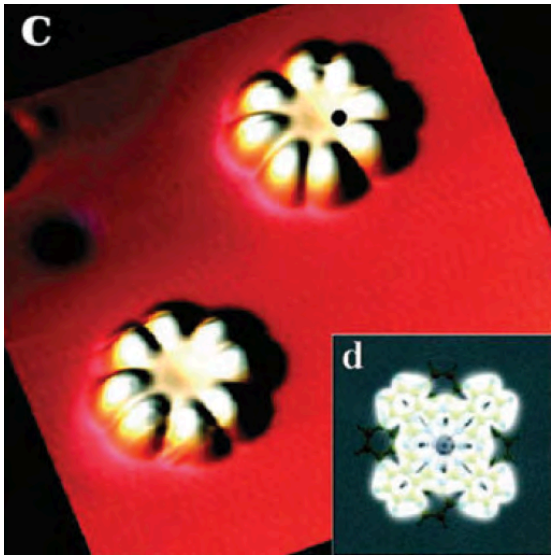


Molecular spin quantum technology?

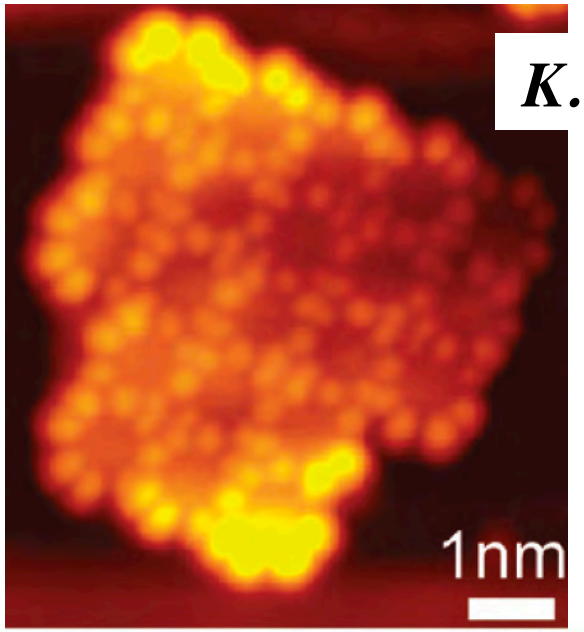


Organisation on surfaces

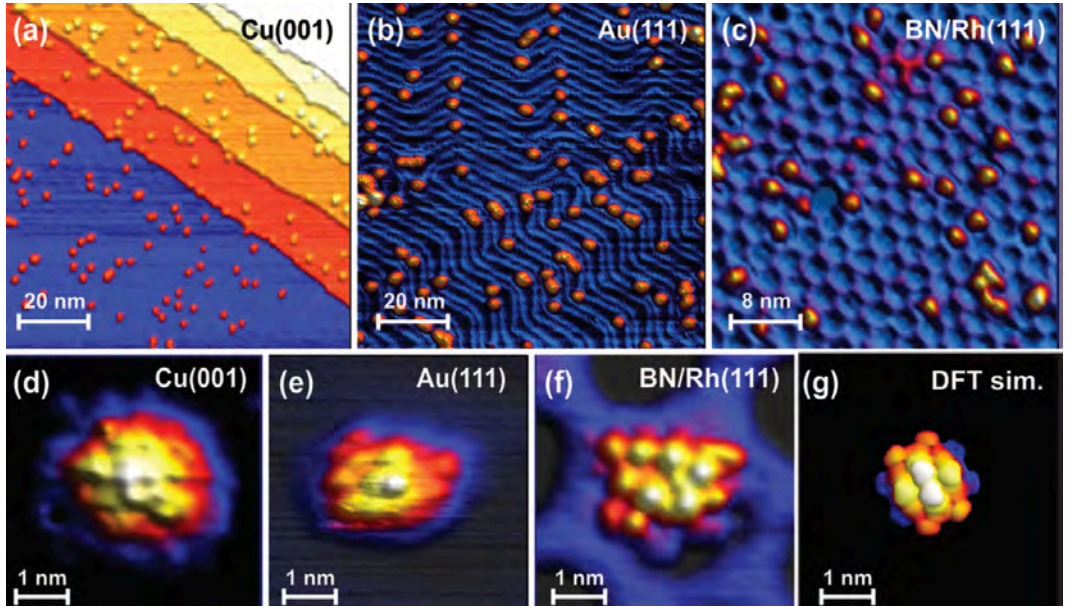
A. Cornia



K. Kern



K. Katoh



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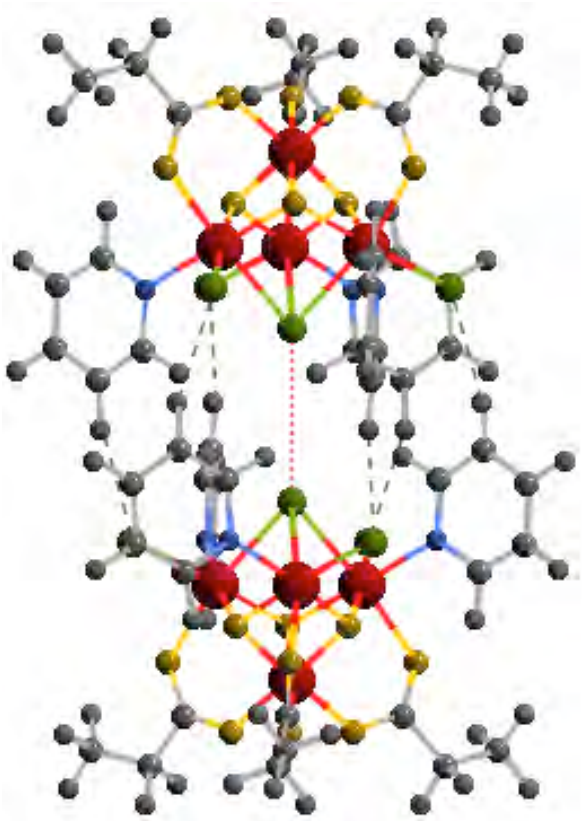
  T_1 up to years, T_2 ~ μs – ms
- Peripheral functionalization of SMMs
- Organisation on surfaces
- Interconnecting SMMs
  Static: exchange-biased
  Switchable: photo-induced electron transfert

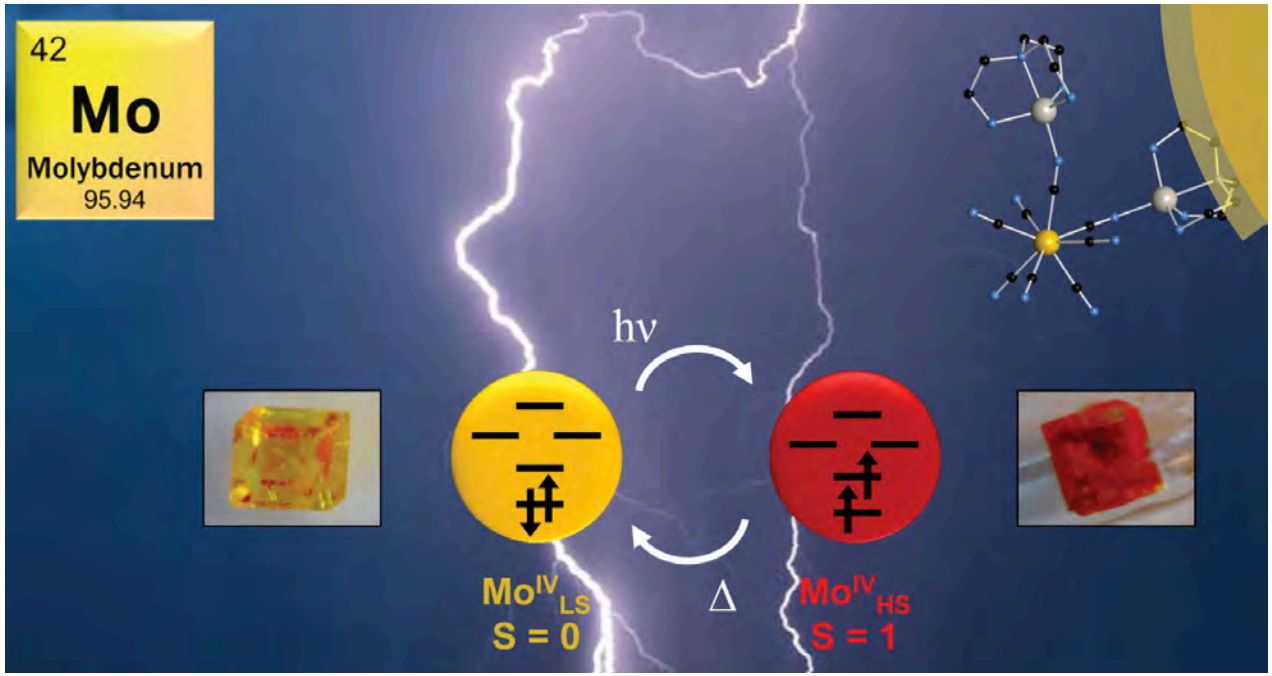


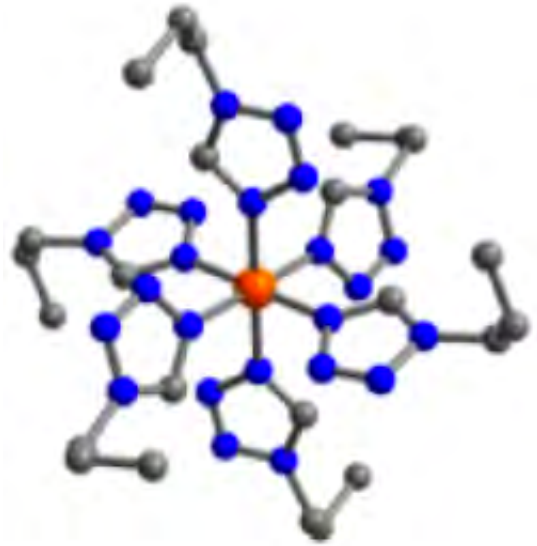
Nature **416**, 406 (2002)

Molecular spin quantum technology?

- **Light-induced spin transition**

[Fe(ptz)6](BF4)2



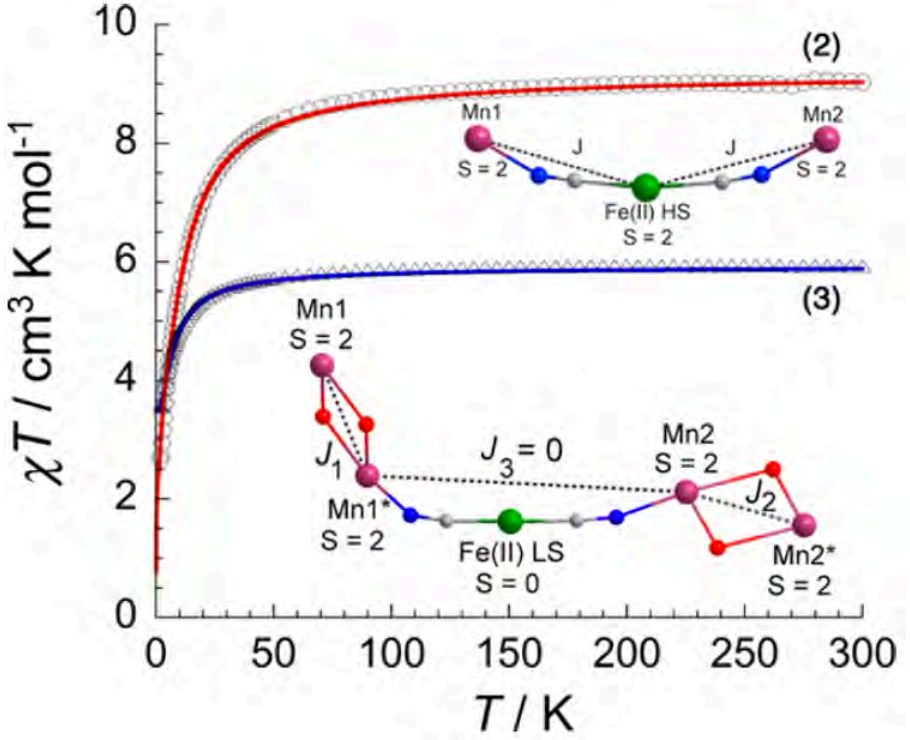


Chem. Commun. 51, 8229 (2015)

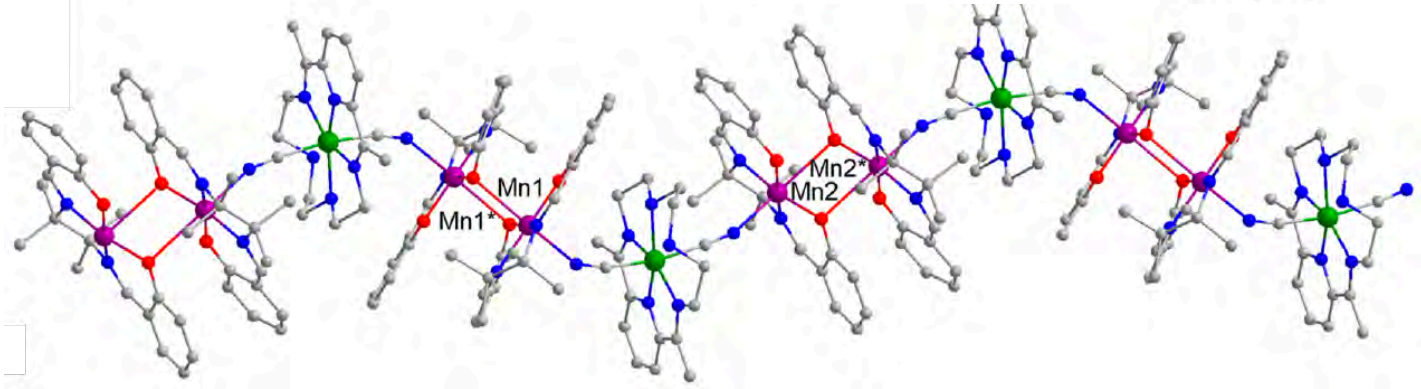
JACS 135, 15880 (2013)

Molecular spin quantum technology?

- **Photo-switchable linkers**



JACS 135, 14840 (2013)



Molecular spin quantum technology?

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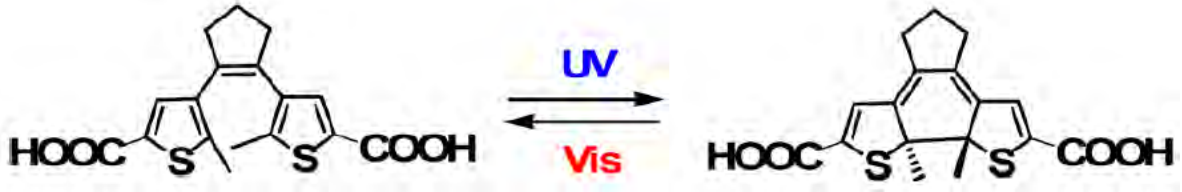
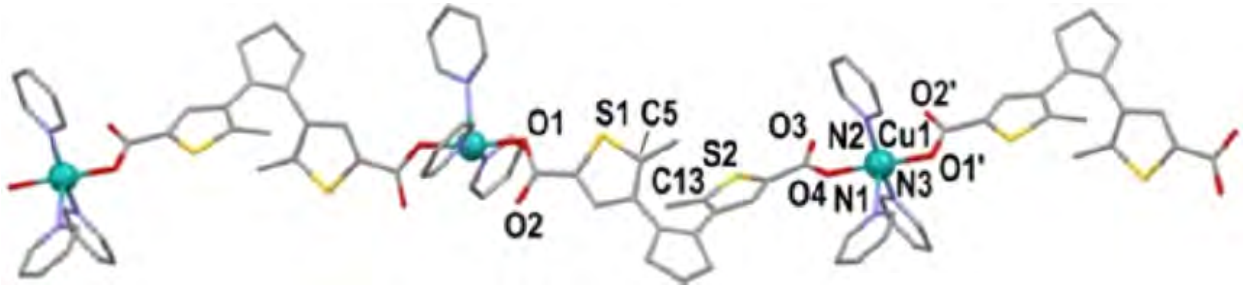
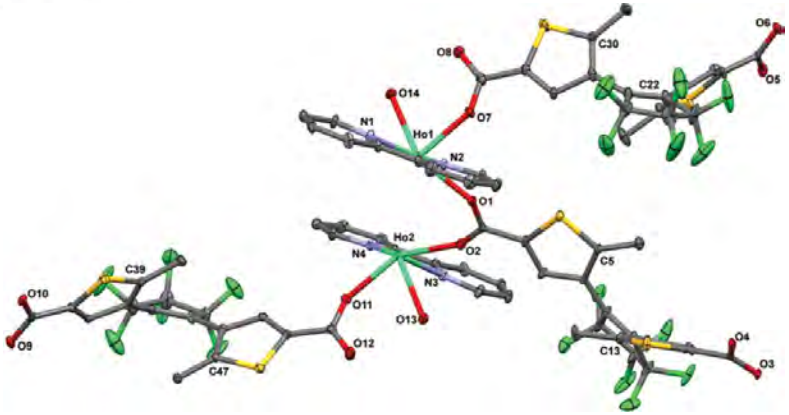


Photochromic DTE ligand with two carboxylic groups in the open and closed form

J. Am. Chem. Soc. 122, 7195 (2000)



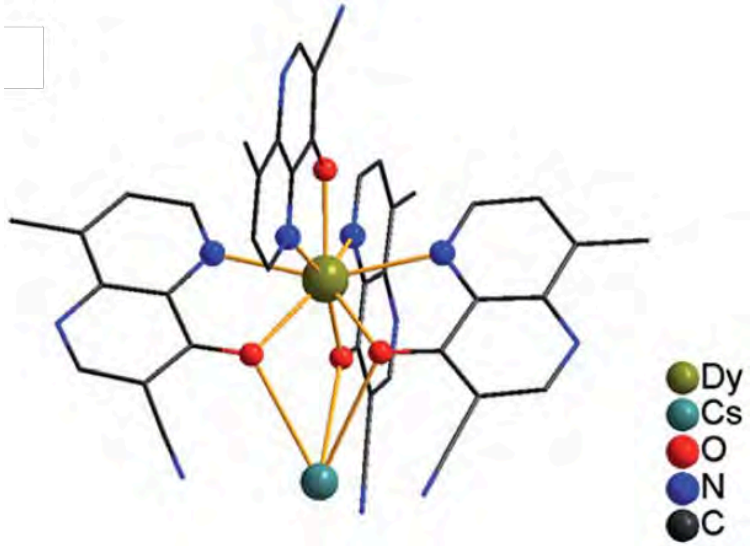
Cryst. Growth Des. 16, 4026 (2016)

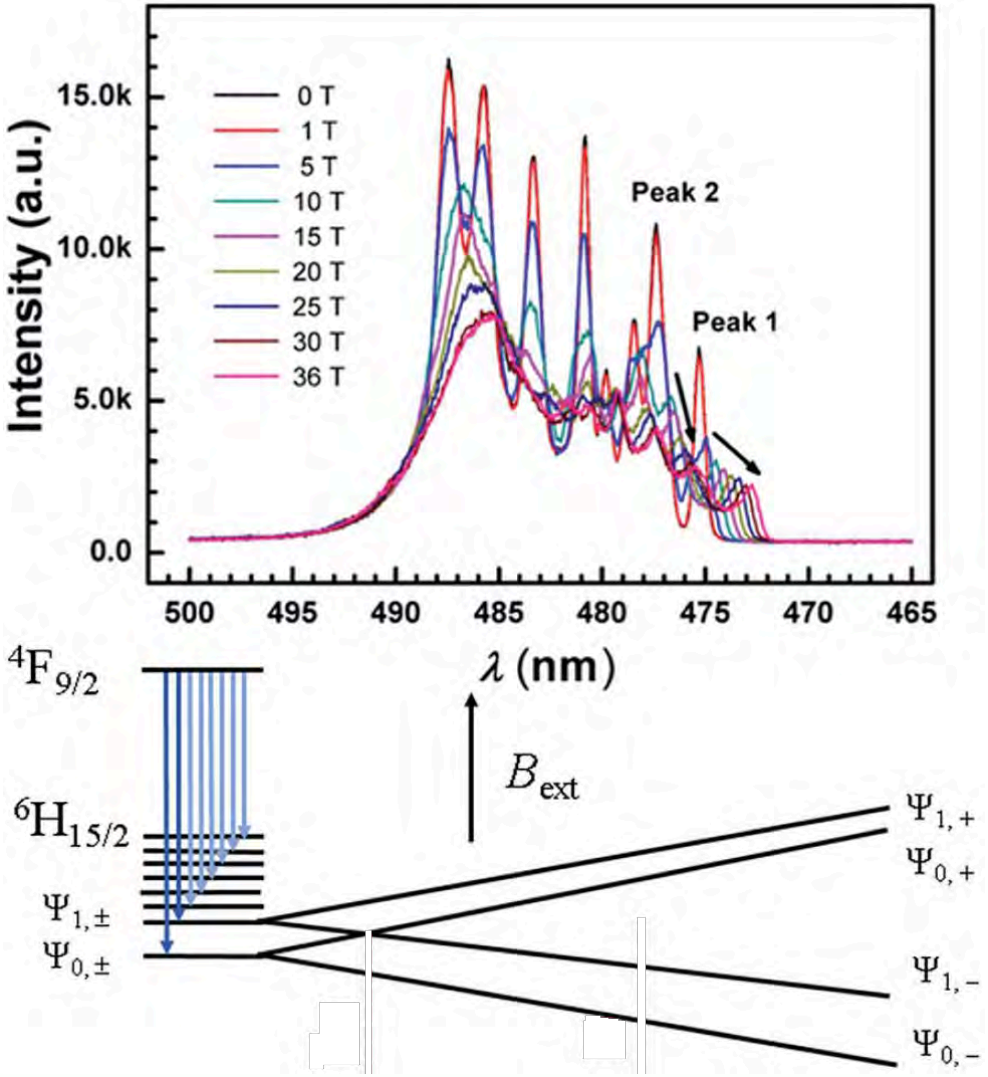


Dalton Trans. 44, 5996 (2015)

Molecular spin quantum technology?

- **Photoluminescence for read-out**

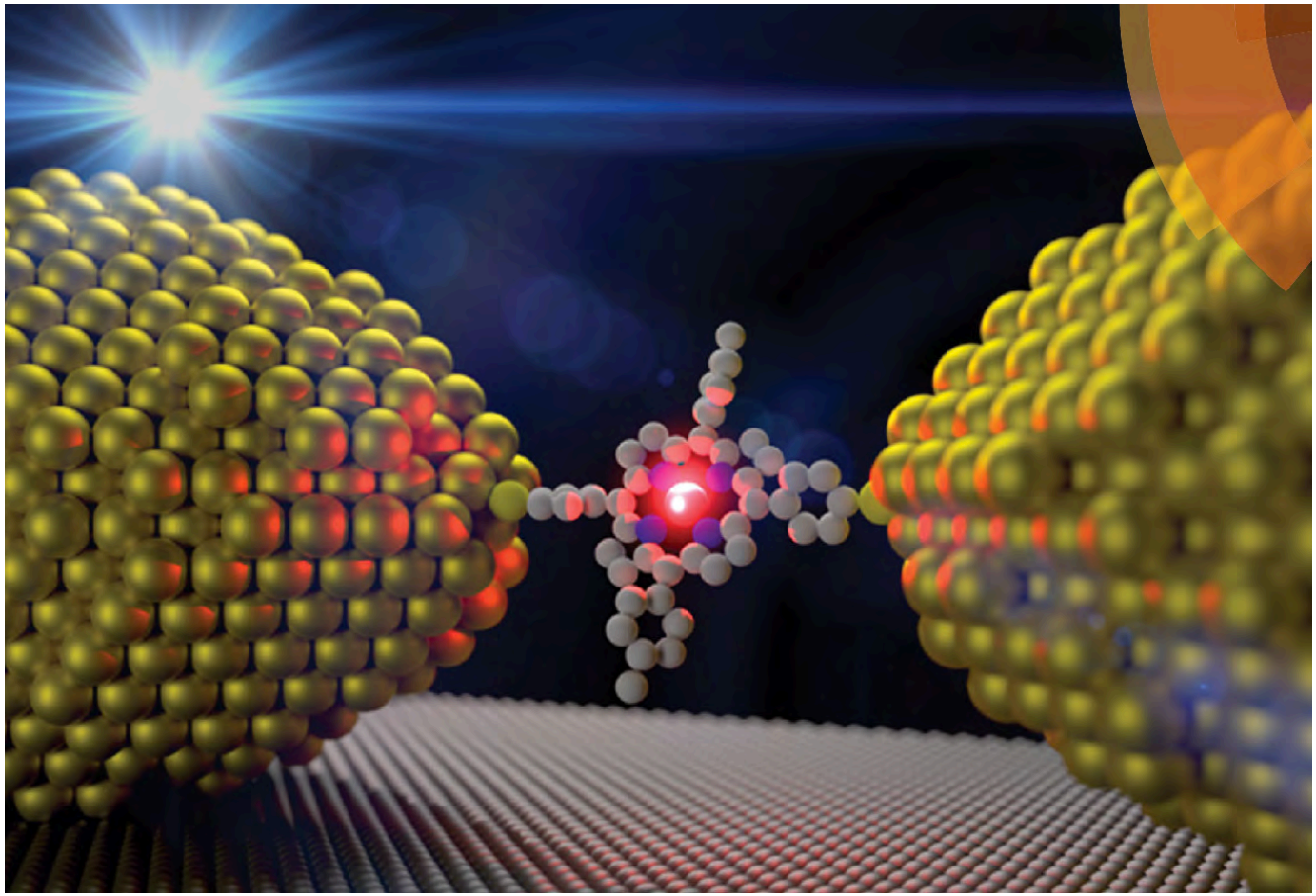




Chem. Sci., 7 5020 (2016)

Molecular spin quantum technology?

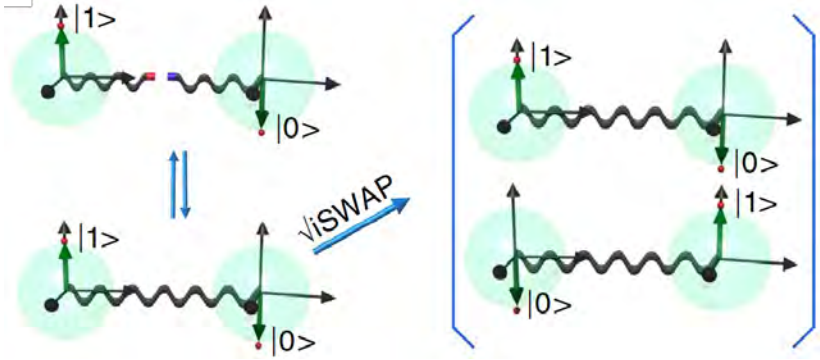
- **Molecular quantum devices**

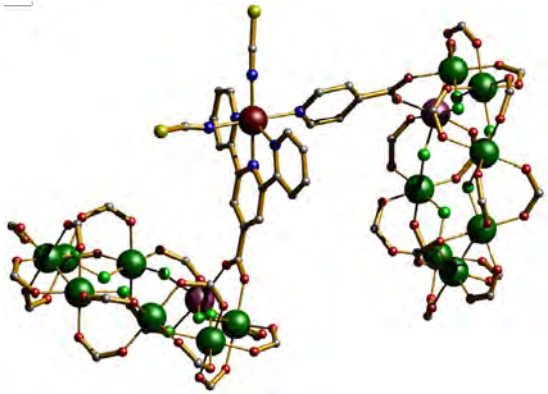


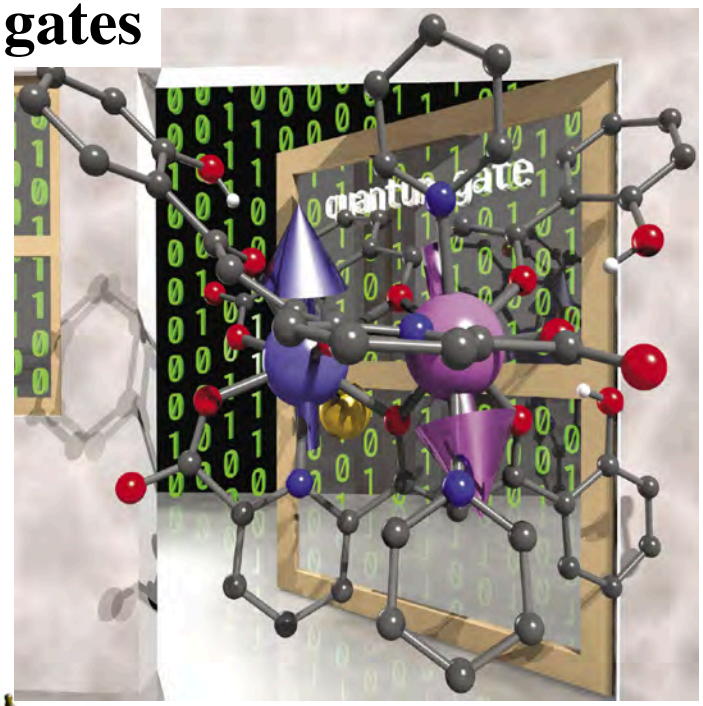
M.L. Perrin, E. Burzuri, and H.S.J. van der Zant
Chem. Soc. Rev. 44, 902 (2015)

Molecular spin quantum technology?

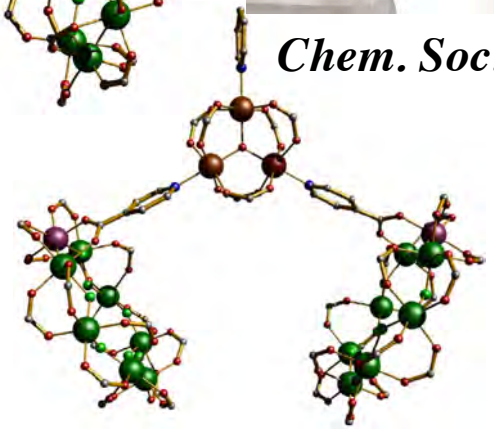
- **Implementation of universal quantum gates**







Chem. Soc. Rev. 41, 537 (2012)

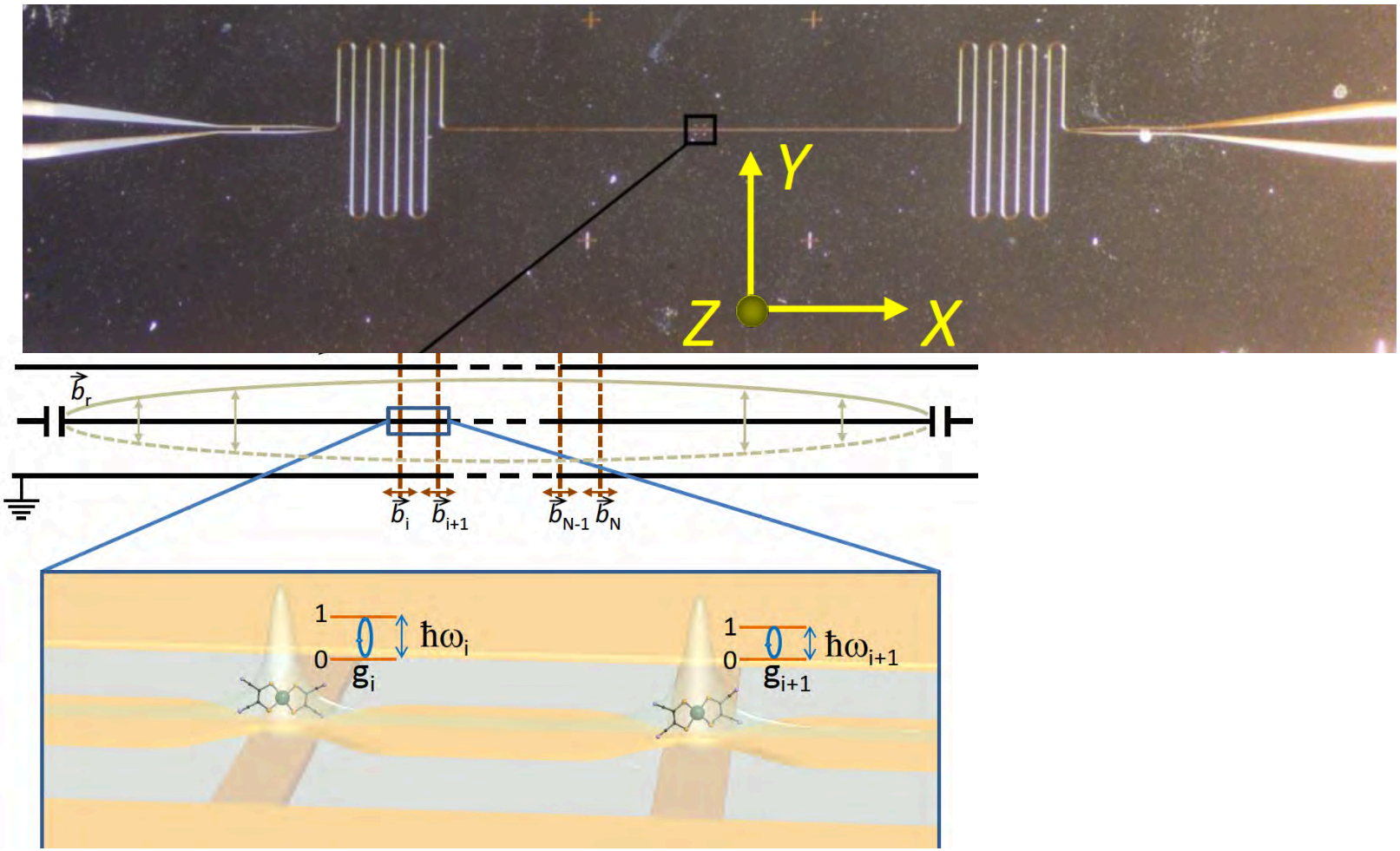


PRL 107, 117203 (2011)

Nature Comm. 7, 11377 (*2016*)

Molecular spin quantum technology?

- **A scalable architecture for quantum computation**



Dalton Transactions (2016) *DOI: 10.1039/C6DT02664H*

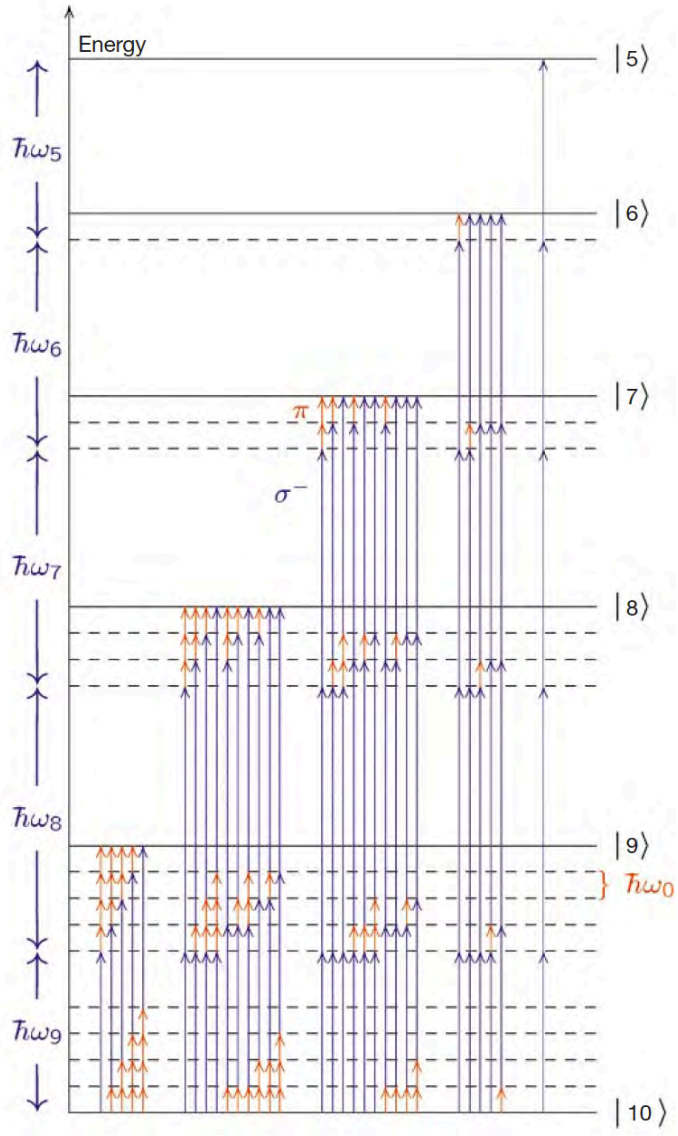
Molecular spin quantum technology?

- **Theory**

Quantum computing in molecular magnets
M.N. Leuenberger & D. Loss
Nature 410, 789 (2001)

> 1400 citations

etc.



Review:
A. Ghirri, F. Troiani, M. Affronte,
Quantum Computation with Molecular Nanomagnets: Achievements, Challenges, and New Trends.
Struct. Bond.164, 383 (2015).

Molecular spin quantum technology?

Quantum computing in molecular magnets
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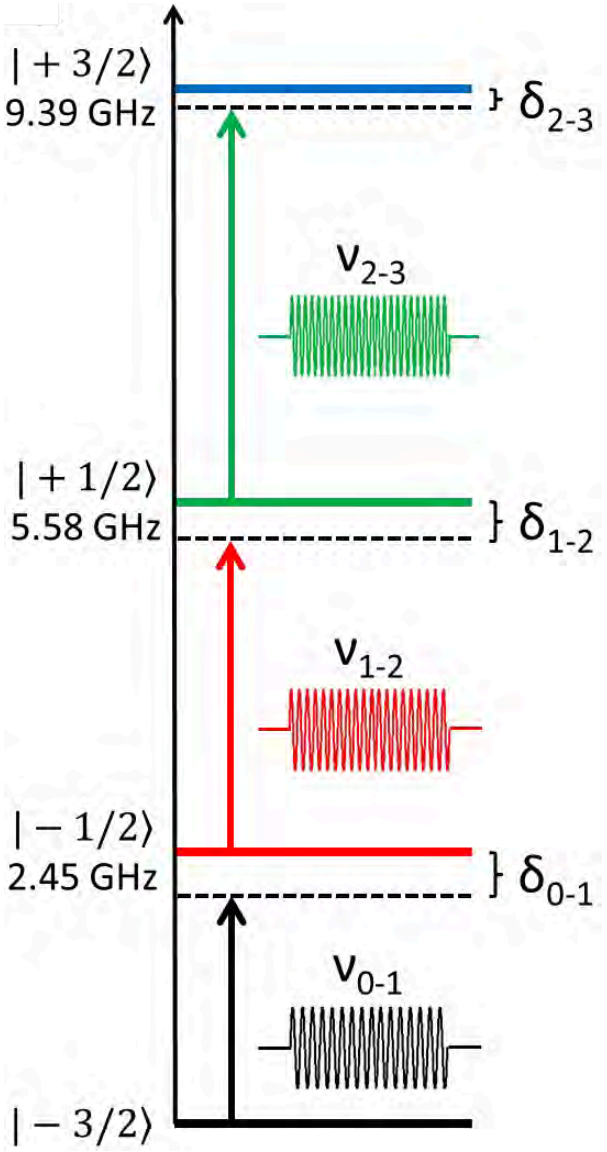
Grover algorithm for large nuclear spins in semiconductors
M.N. Leuenberger & D. Loss
Physical Review B 68, 165317 (2003).

Grover's algorithm



Outline

Di Vincenzo 1996

Isolation

Read-out

Initialization

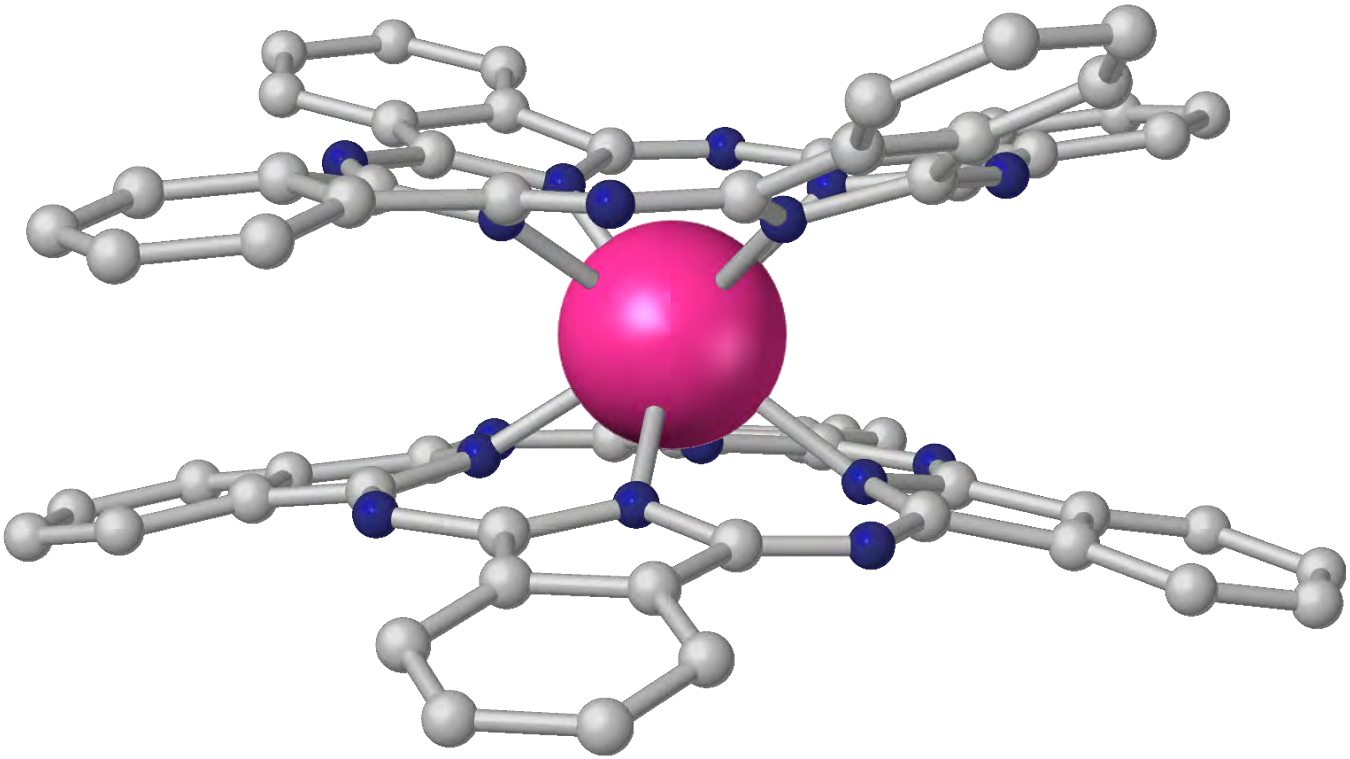
Manipulation

Storage

Grover's algorithm

Nuclear spin-qubit SMM

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



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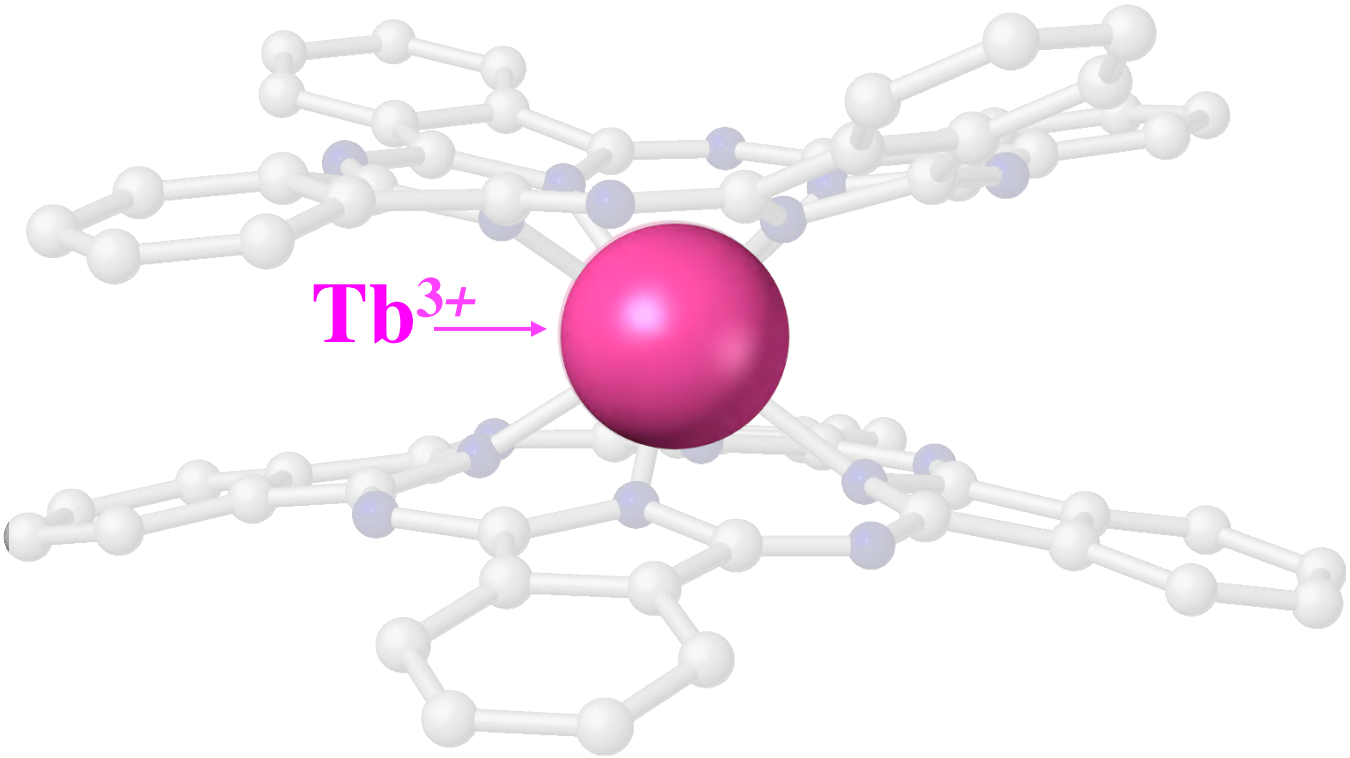


Nuclear spin-qubit SMM

- **terbium double-decker ($TbPc_2$)**

  electronic spin

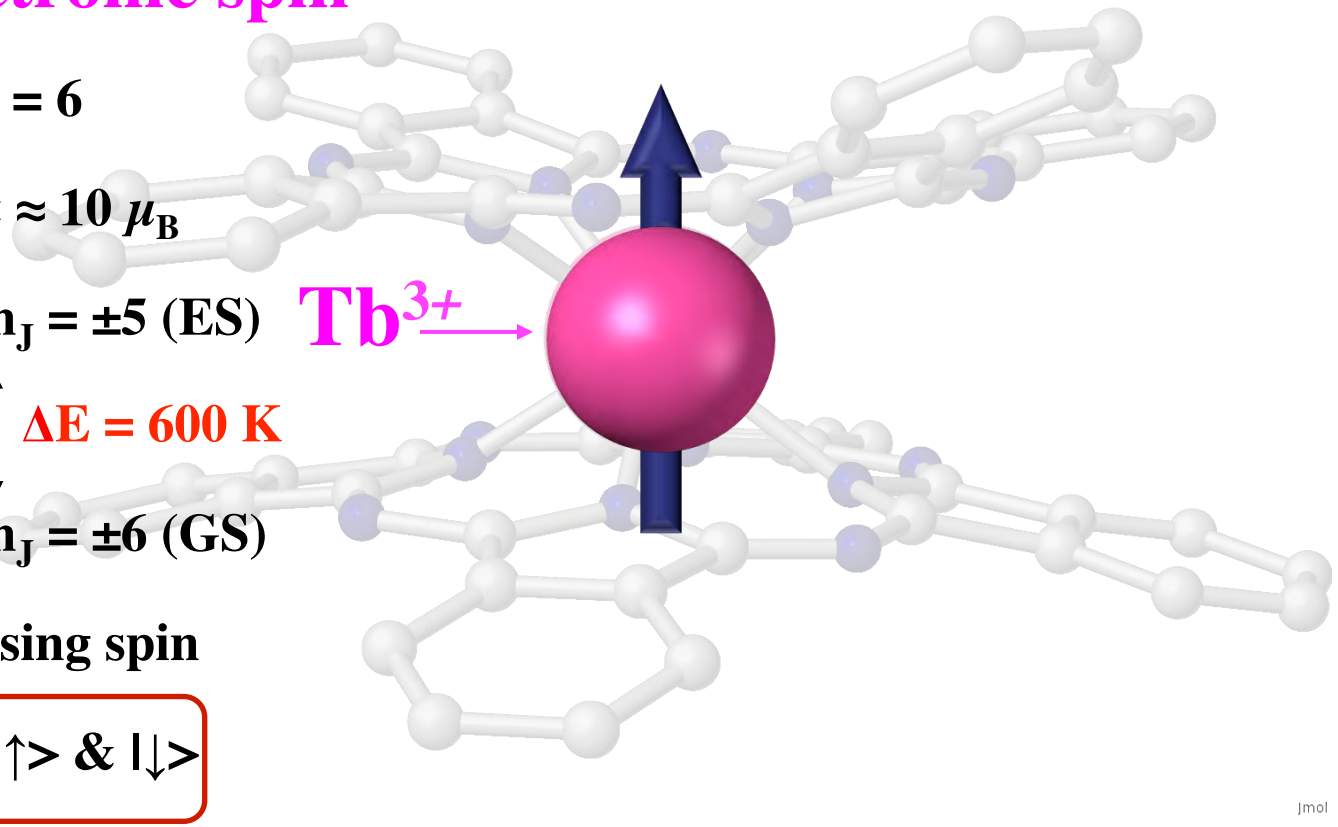
  - **J = 6**
  - **$\mu \approx 10\ \mu_B$**
  - **$m_J = \pm 5$ (ES)**

    $\Delta E = 600$ K

    $m_J = \pm 6$ (GS)

  → Ising spin

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



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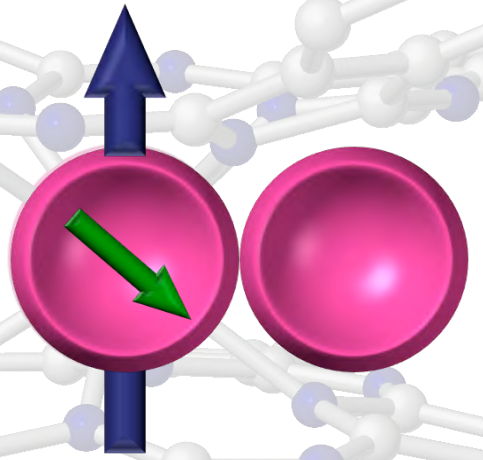
→ Ising spin

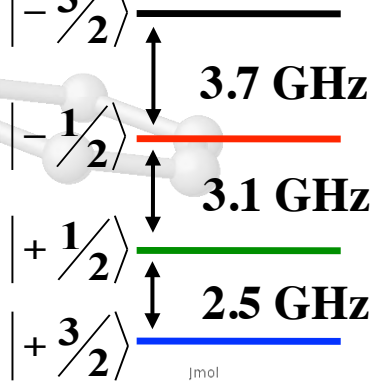
$|\uparrow\rangle$ & $|\downarrow\rangle$



environmental isolation!

nuclear spin

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



Outline

Di Vincenzo 1996

Isolation

Read-out

Initialization

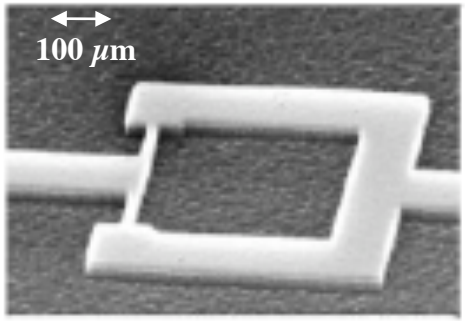
Manipulation

Storage

Grover's algorithm

SMM detectors

- **tiny magnetic moment (few μ_B) → sensitive detectors**

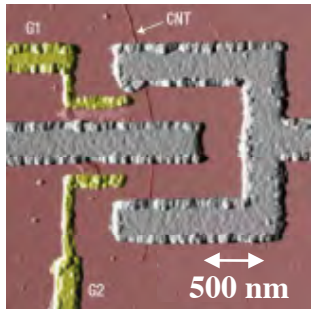


Phys. Rev. **Lett. 77, 1873** (1996)

sensitivity :
10^2 - 10^3 spins

size reduction →

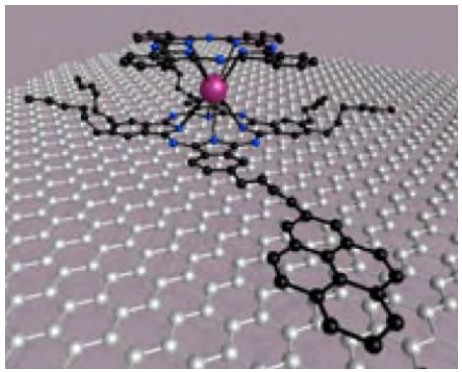
Nano-SQUID



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

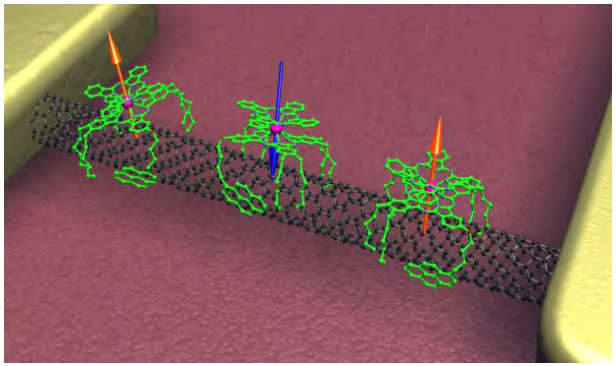
↓ **different concepts**

2D



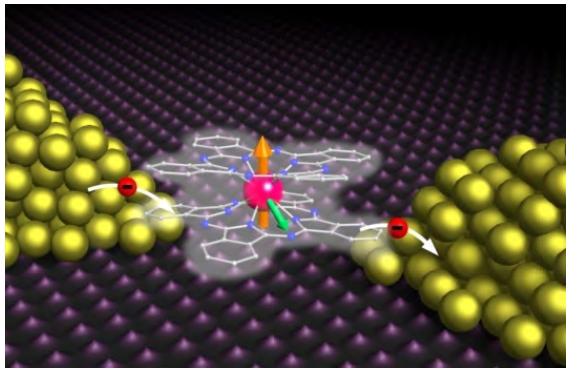
Nano Lett. **11, 2634** (2011)

1D



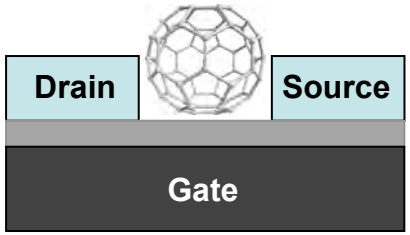
Nature Materials **10**, 502 (2011)

0D



Nature **488**, 357 (2012)

Fabrication of a single-molecule transitor

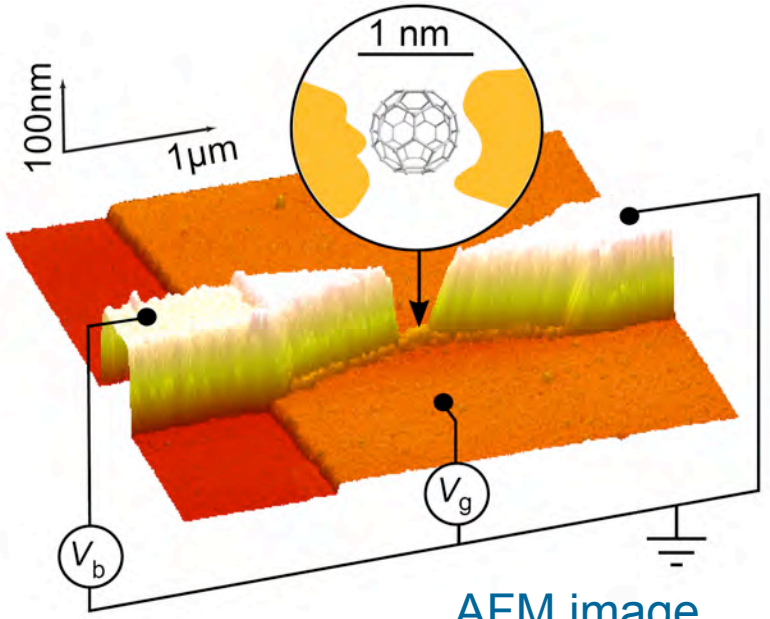


Electromigration:

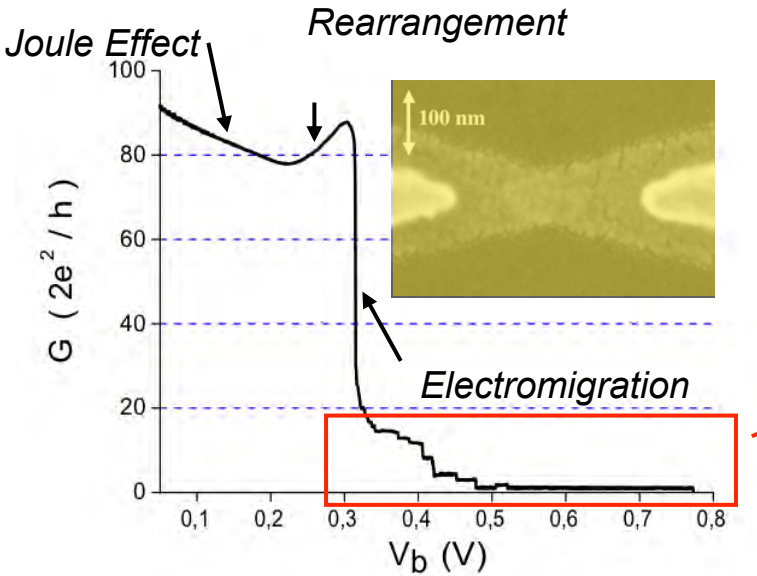
- Park et al, APL, **75**, 301-303 (1999)
- D.R. Strachan et al, APL, **86**, 043109 (2005)
- Zheng Ming Wu et al, APL, **91**, 053118 (2007)
- K. O' Neill et al, APL, **90**, 133109 (2007)

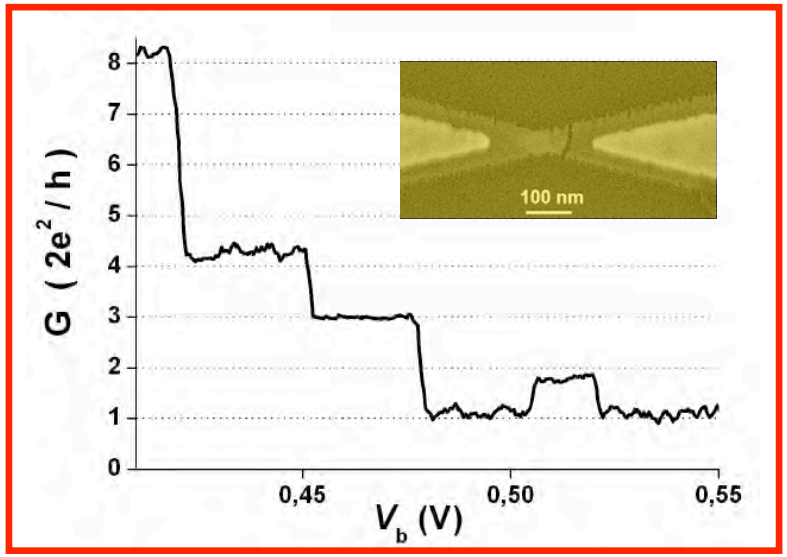
Set-up:

- Dilution fridge 35 mK
- Low temperature filtering
- Fast feedback ≈ 1.5 μs

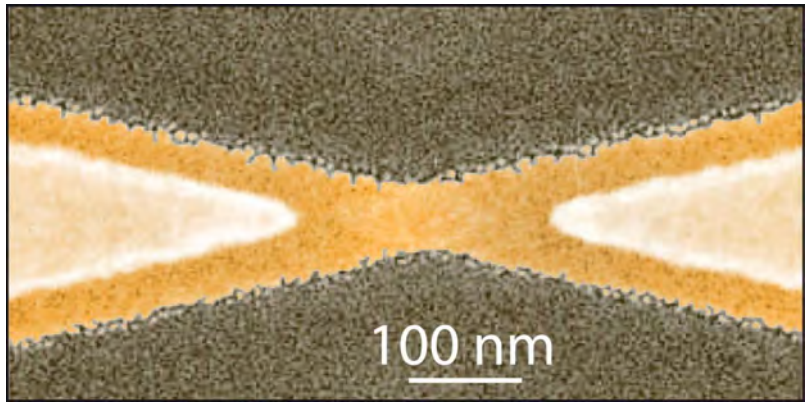


AFM image

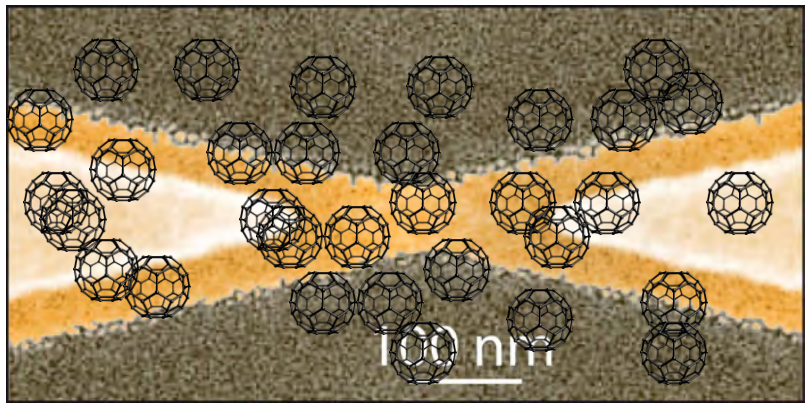




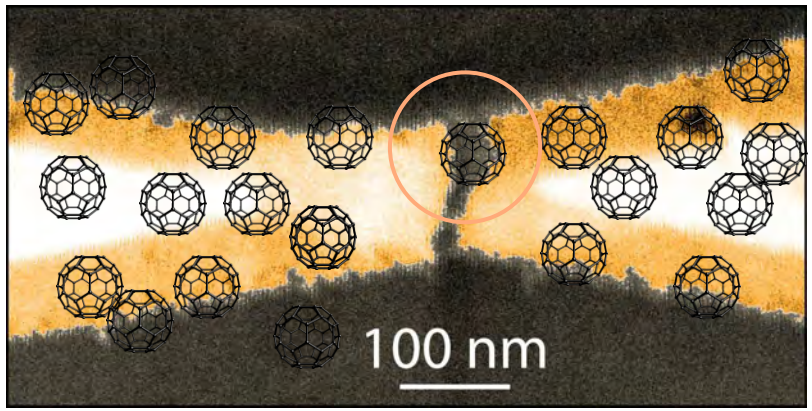
Fabrication of a single-molecule transitor



Nano-wire



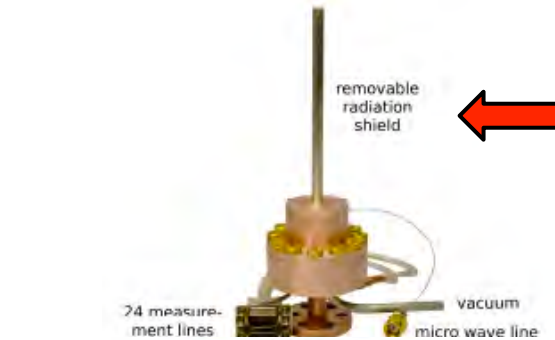
Molecule deposition: blow drying a solution

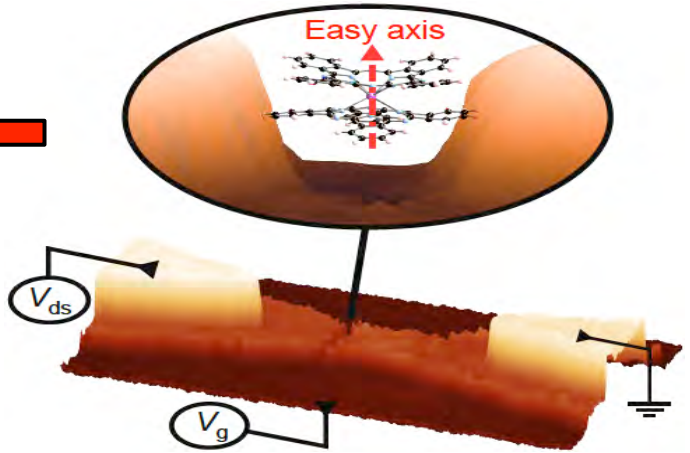


Electromigration and diffusion

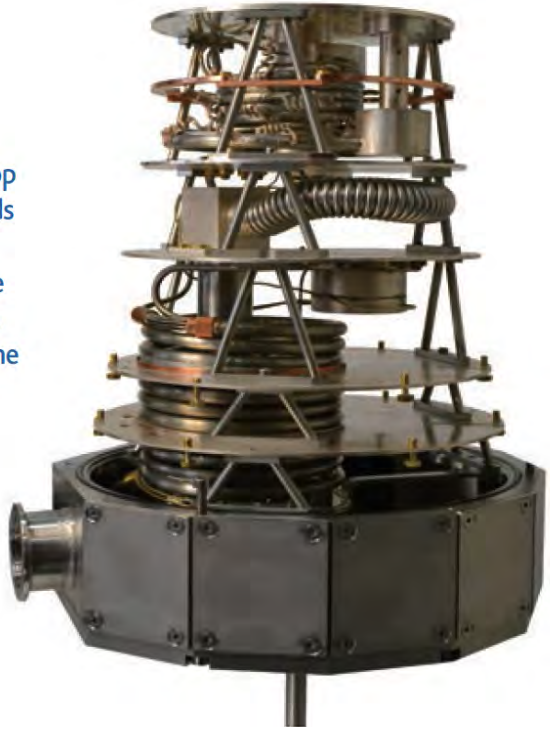
Main problem : yield …

Table-top dilution cryostat: Sionludi



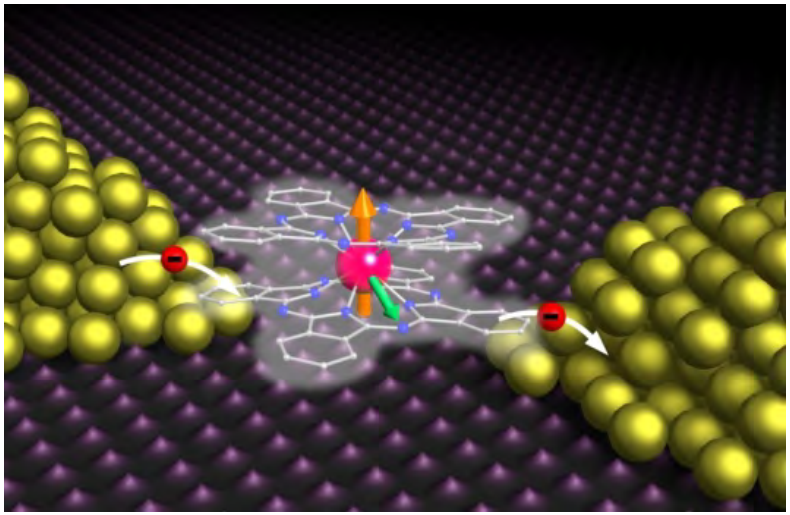


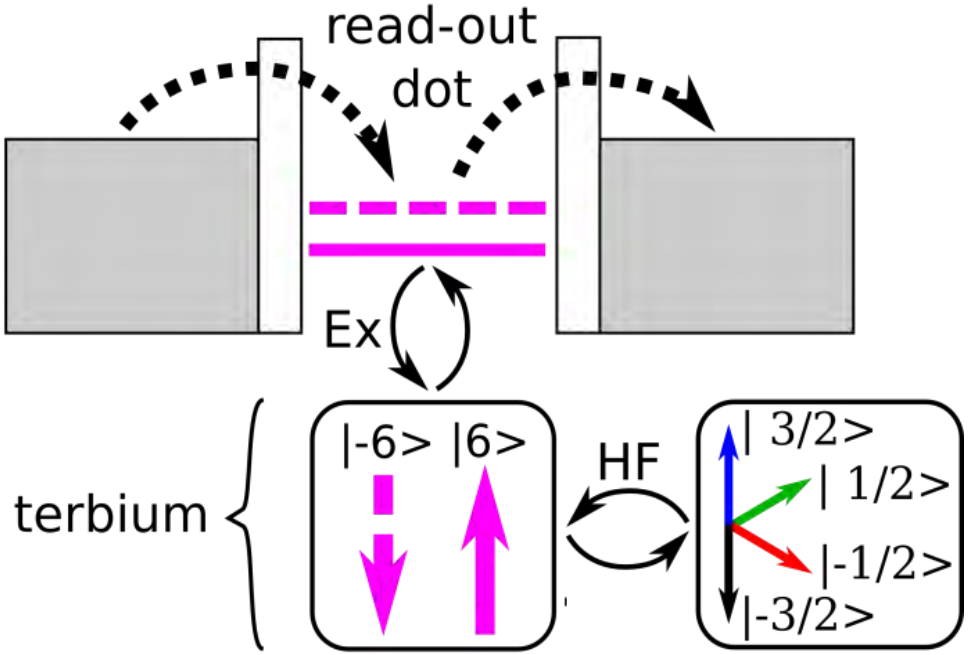
New generation of the Sionludi table-top dilution cryostat, with its thermal shields and outer vacuum container removed. The compact, inverted design allows the cryostat to be mounted on a bench top. The sample space is located on top of the upper plate, making it easily accessible.



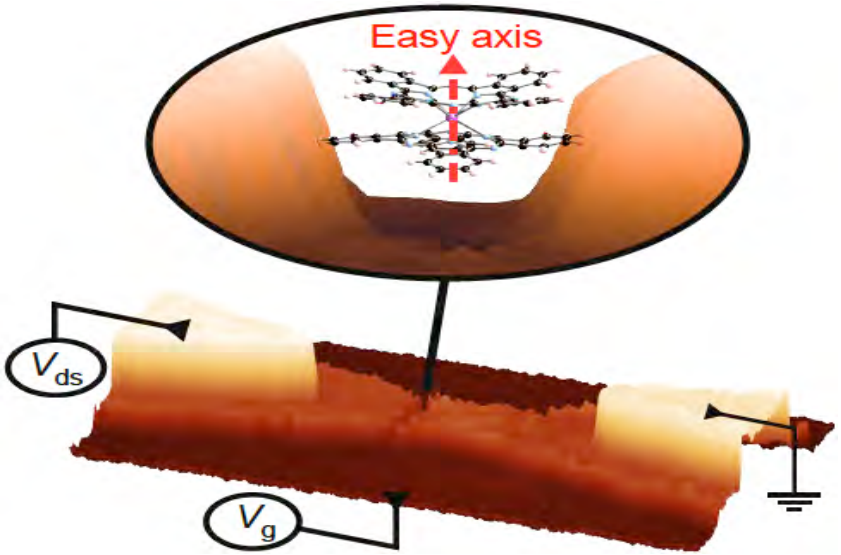
Schematics of the read-out

Weak two-stage-coupling to avoid backaction

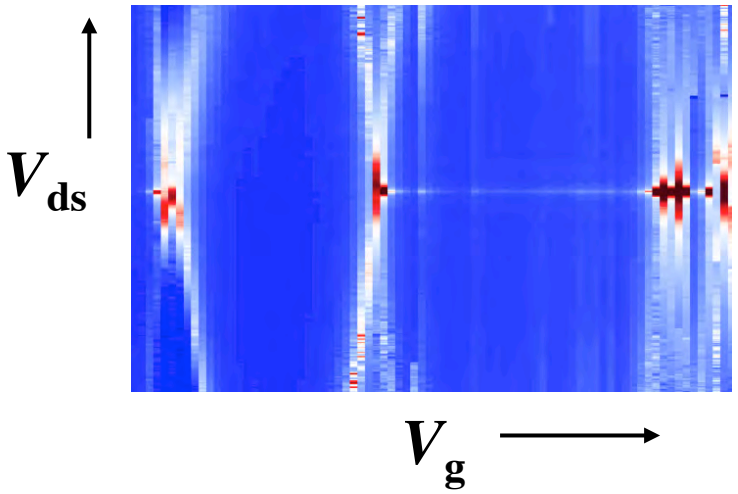


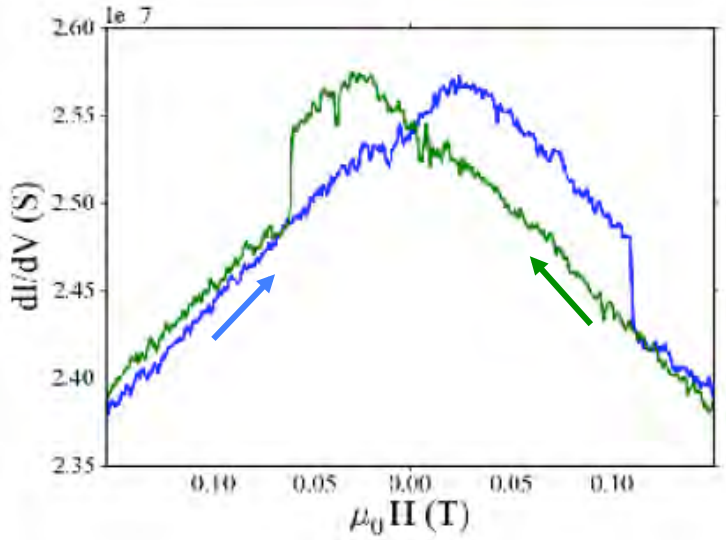


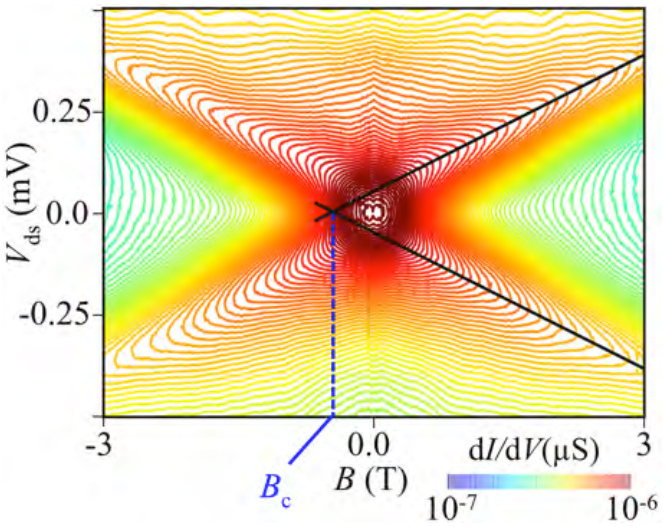
Device characteristics



similar to

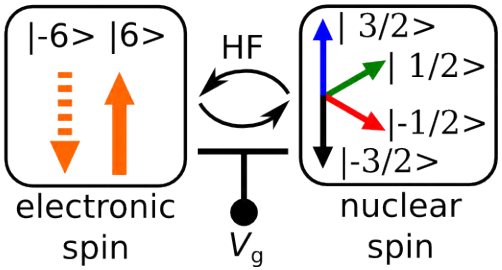


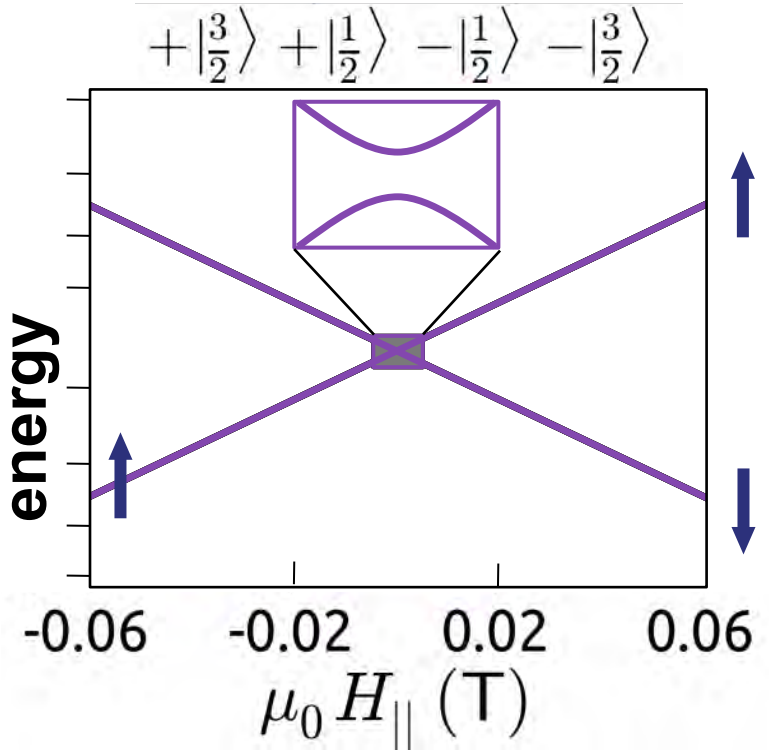




Nuclear spin read-out

- 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

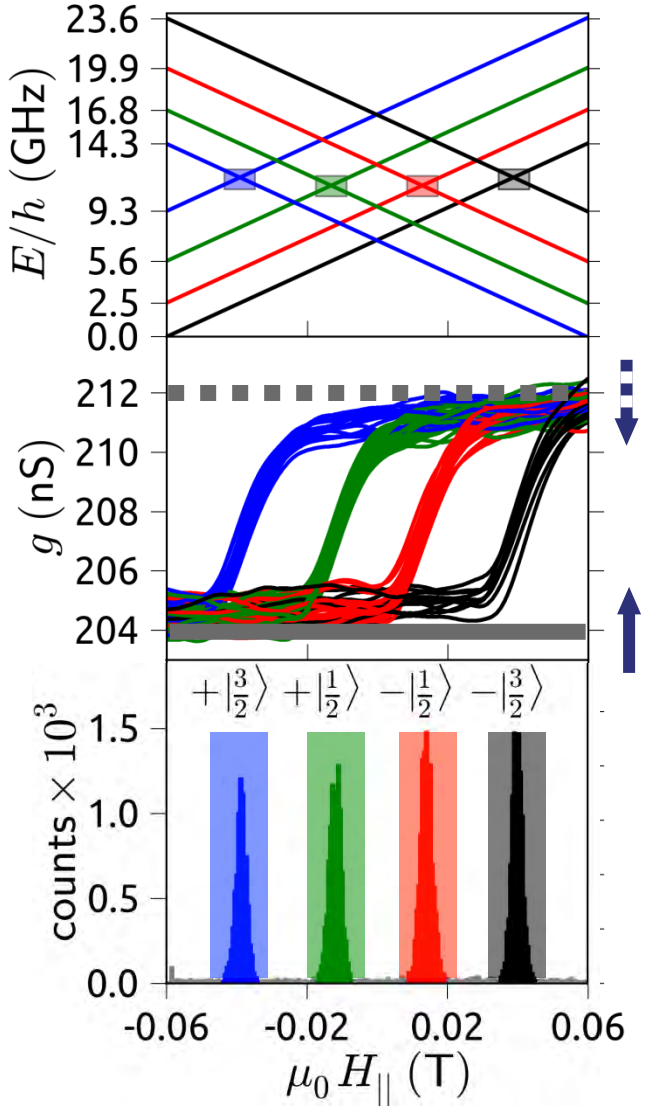
1st nuclear spin → electronic spin

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

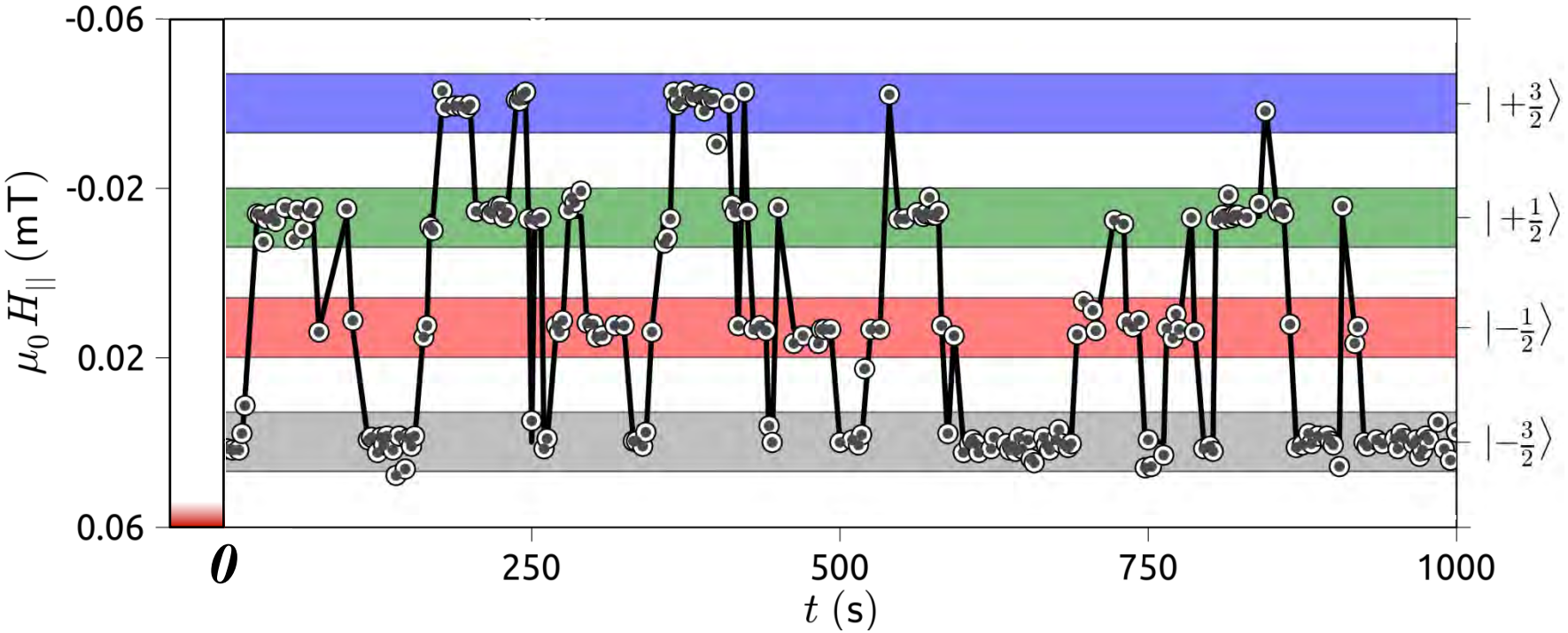
Manipulation

Storage

Grover's algorithm

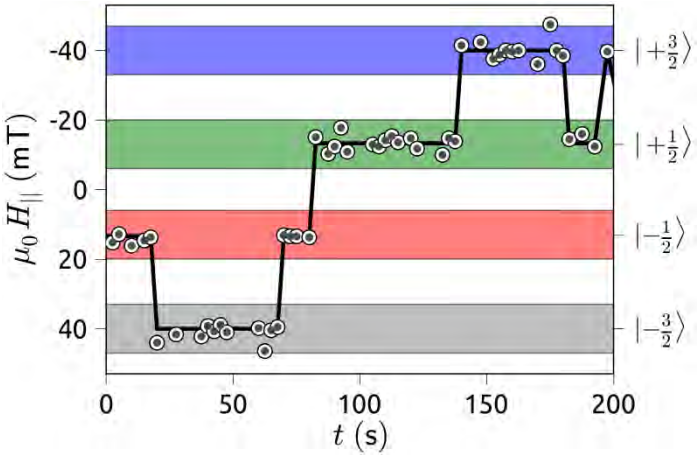
Nuclear spin trajectory

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



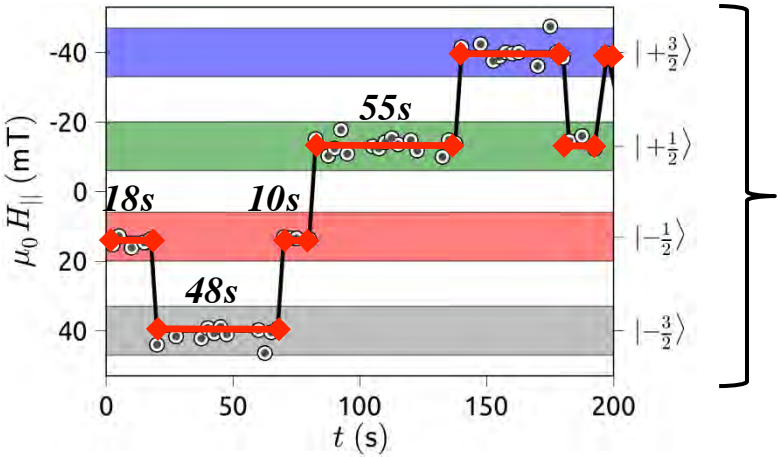
Thiele et al. PRL 111, 3 (2013)

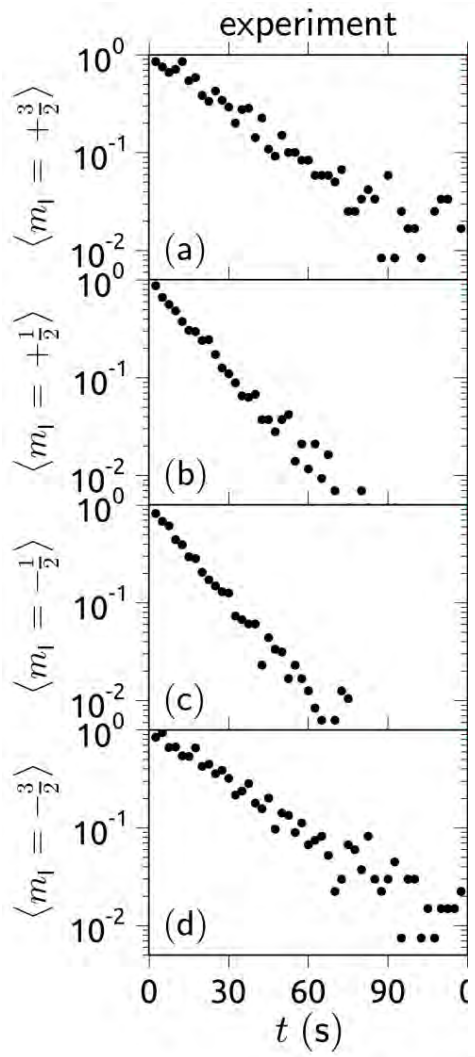
Single nuclear spin relaxation



Thiele et al. PRL 111, 3 (2013)

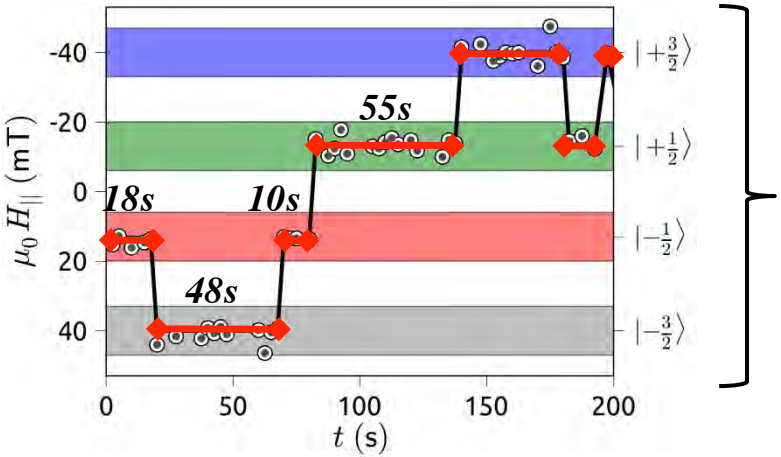
Single nuclear spin relaxation

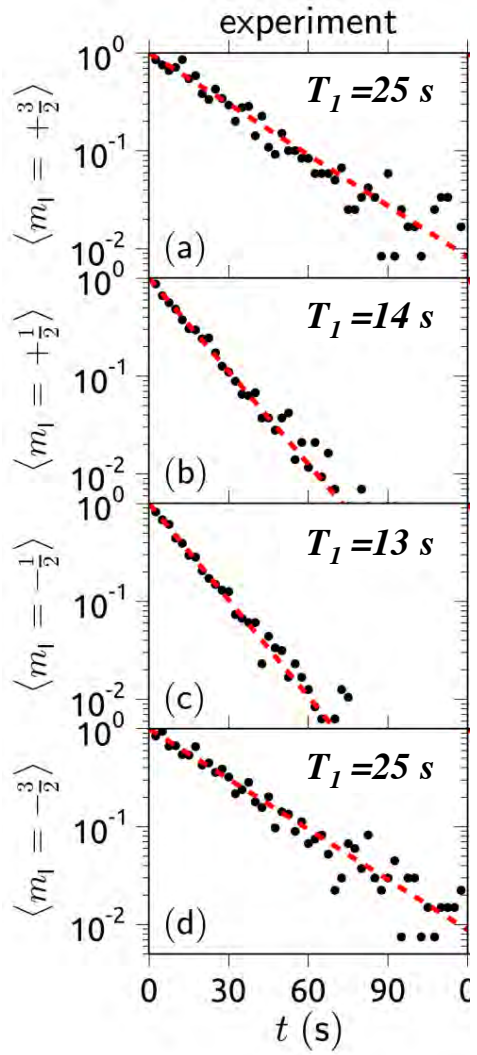




Thiele et al. PRL 111, 3 (2013)

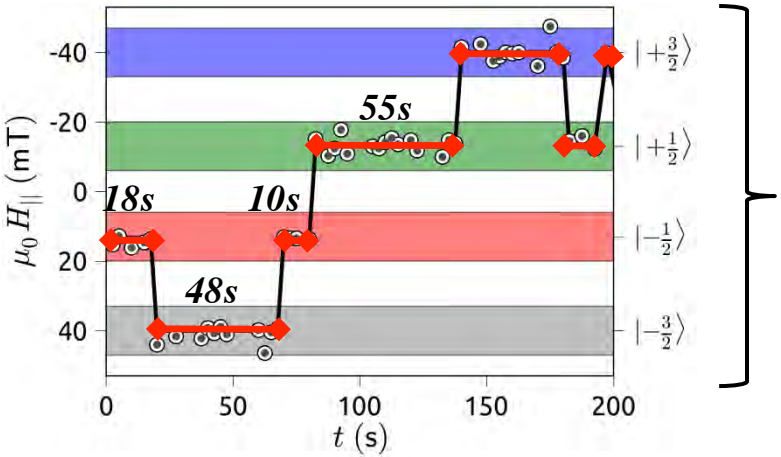
Single nuclear spin relaxation

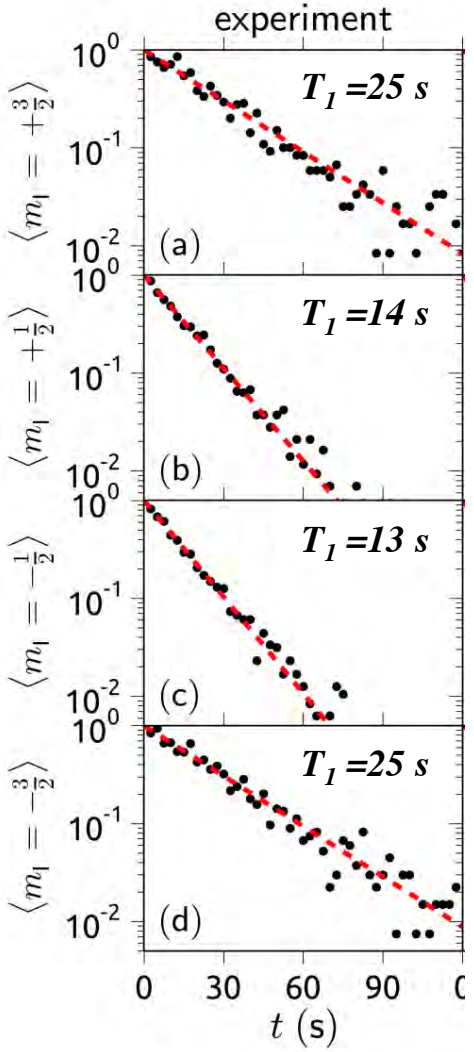




Thiele et al. PRL 111, 3 (2013)

Single nuclear spin relaxation





1 escape path

2 escape paths

2 escape paths

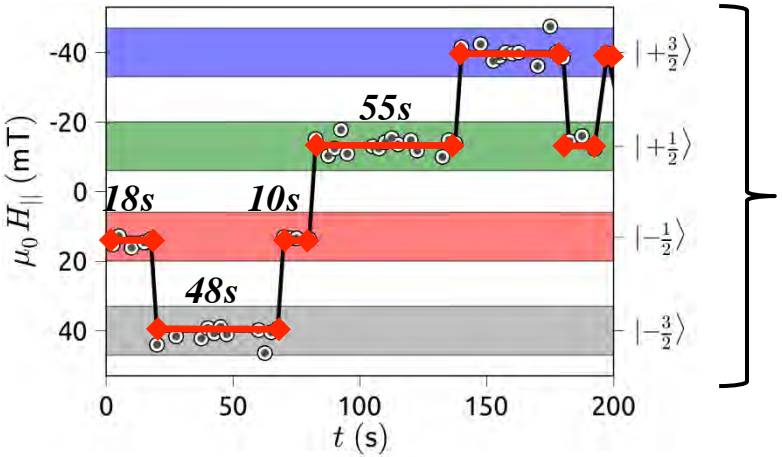
1 escape path

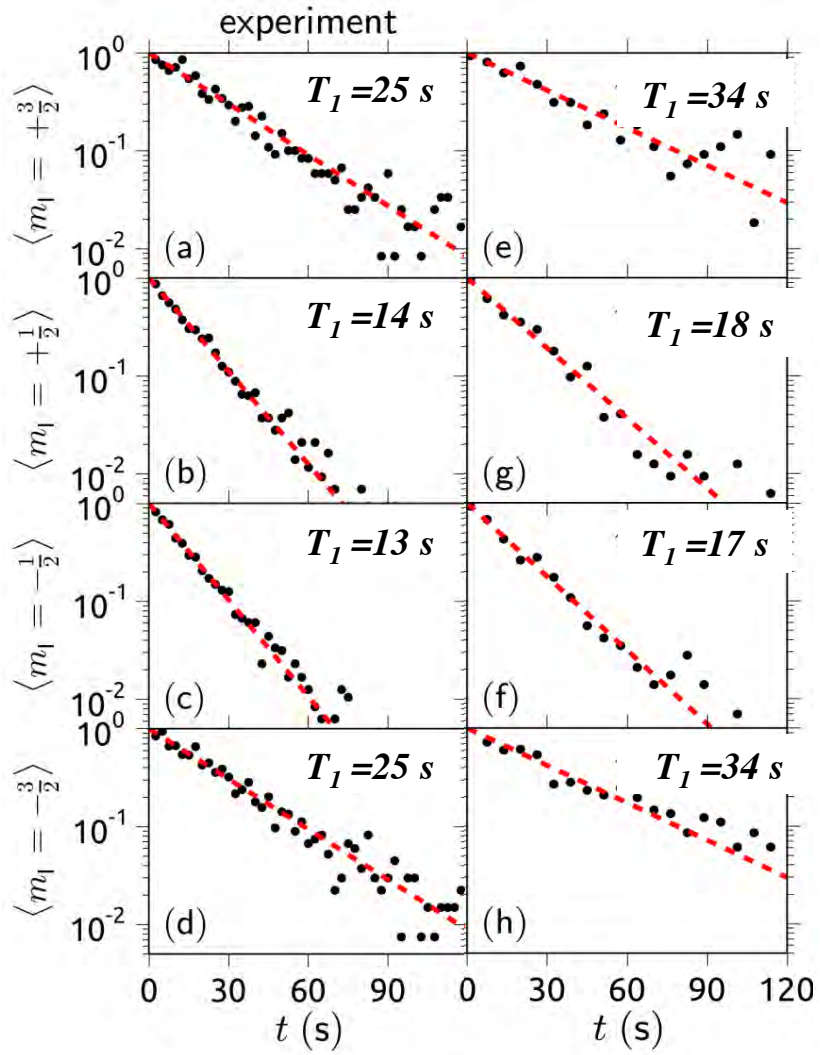
- *plot dwell times in a histogram*
- *fit to $e^{-t/T1}$ → relaxation time $T_1 \approx 10$ s*
- *difference of 2 in T_1 → relaxation path*

→ *measurement = initialization*

Thiele et al. PRL 111, 3 (2013)

Lifetime of the four nuclear spin states





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Thiele et al. PRL 111, 3 (2013)

Outline

Di Vincenzo 1996

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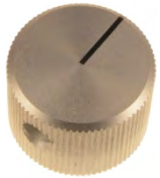


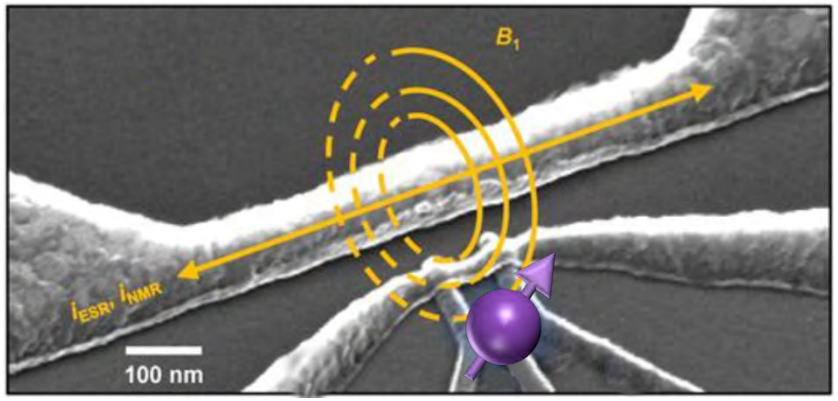
Manipulation

Storage

Grover's algorithm

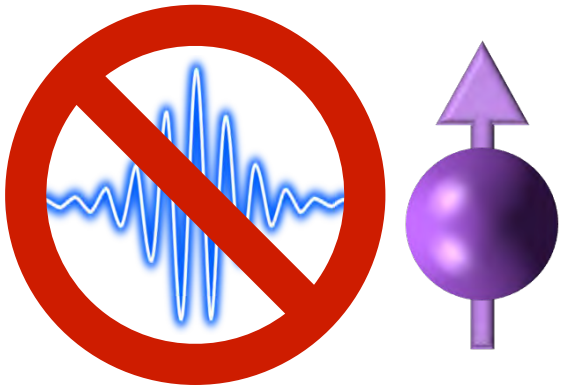
Nuclear spin manipulation

- **using AC magnetic fields**
  - large currents are driven through microwave lines in the vicinity of the sample
  - reliable and straight forward technique
  - BUT:
    - Joule heating
    - parasistic crosstalk
    - only fields up to 3 mT at mK temp.



Pla et al., Nature 496, 334 (2013)

- **using AC electric fields**
  - easy to generate
  - only displacement currents → less Joule heating
  - easy to focus → less crosstalk
  - BUT:
    - no direct interaction between spin and electric field

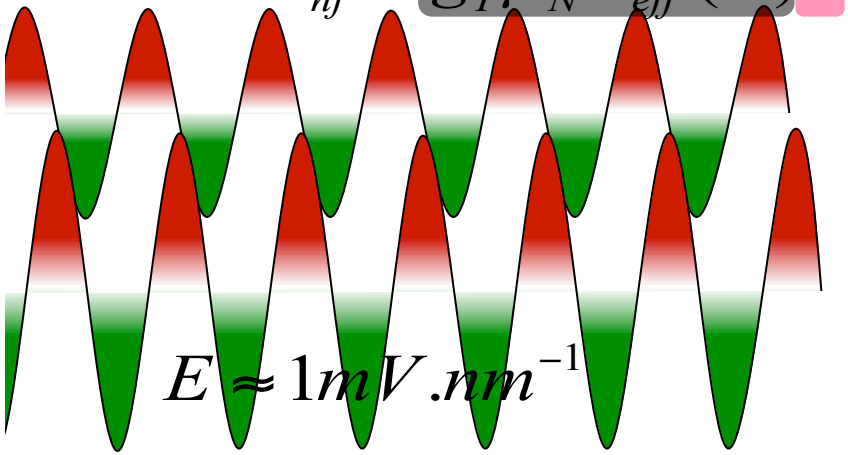


Single Nuclear Spin Manipulation

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

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

Electric field ! Magnetic field ?

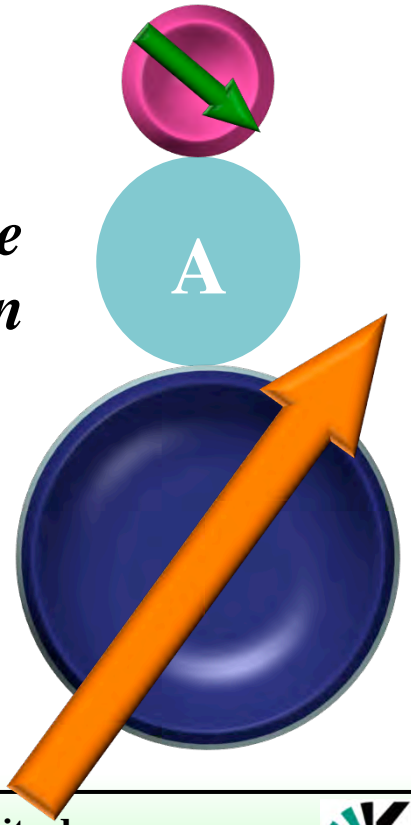


$$E \approx 1 mV.nm^{-1}$$

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

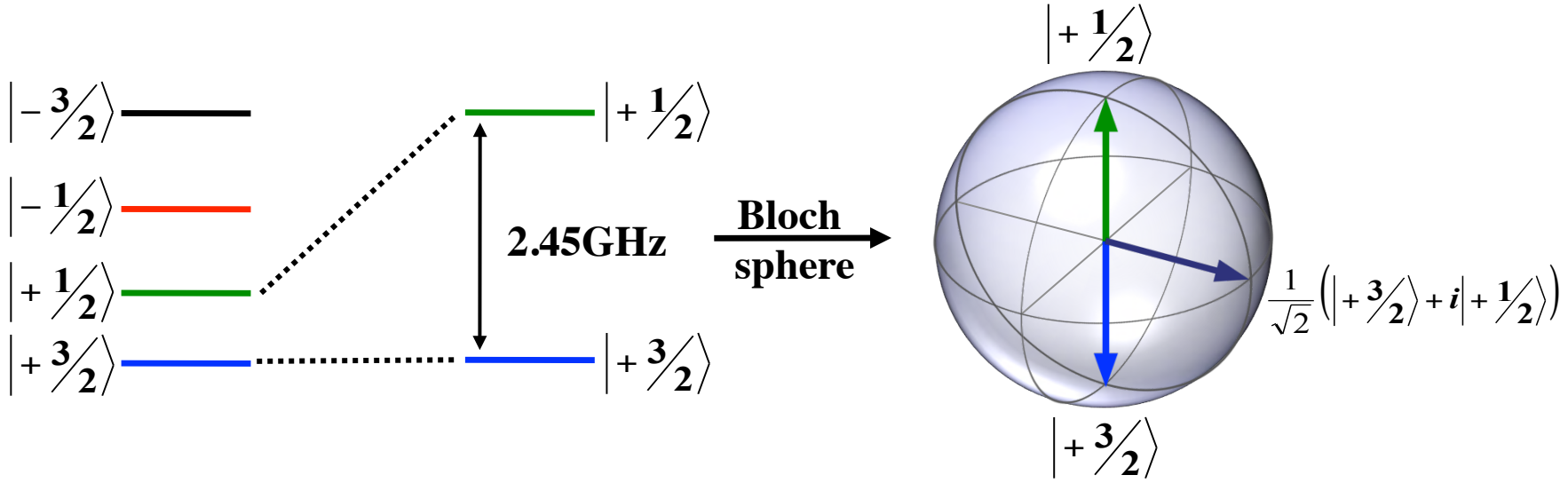
$$B_{eff} \approx 100 mT$$

Hyperfine interaction



Nuclear spin manipulation

- Due to unequal energy spacing → two level subsystem



Nuclear spin manipulation

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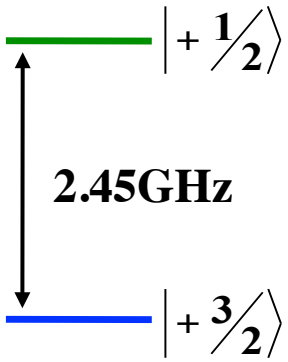
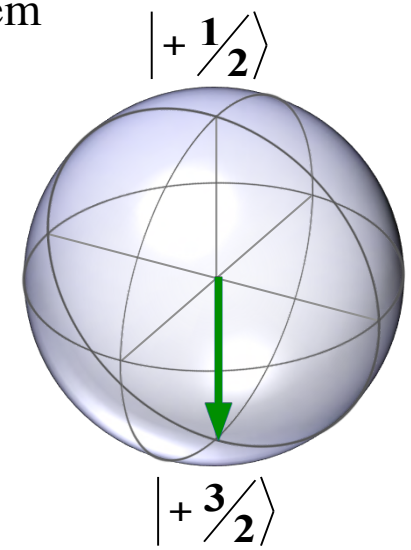
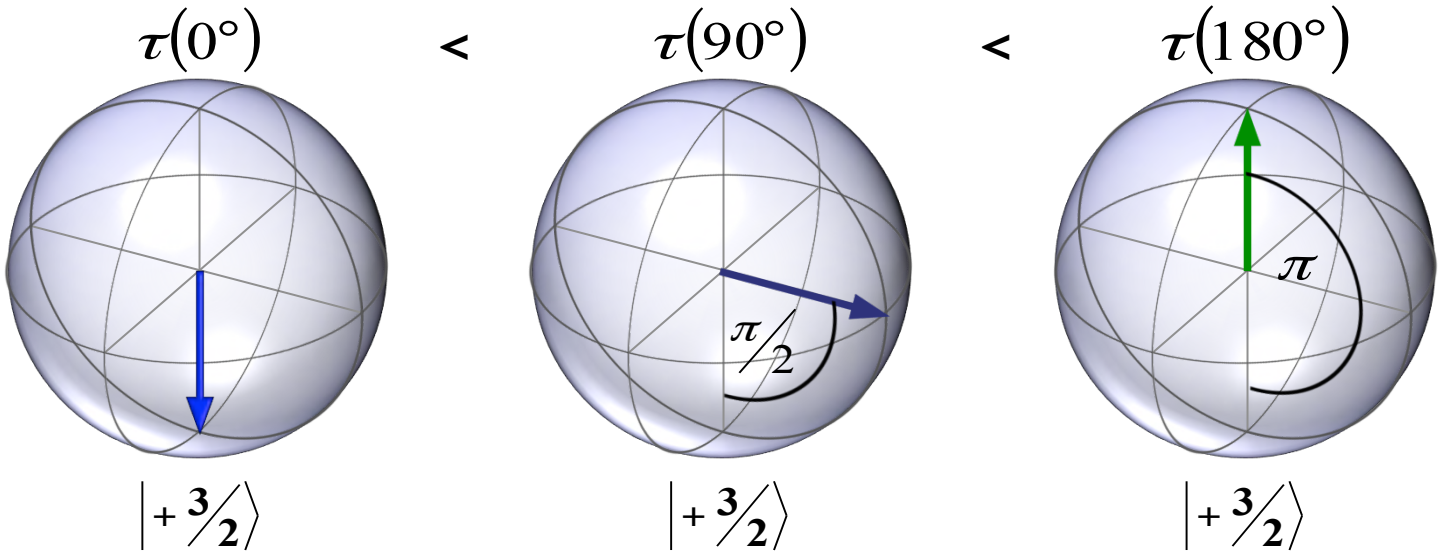




- applying a microwave pulse with f = 2.45 GHz will rotate the vector

Nuclear spin manipulation

- due to unequal energy spacing → two level subsystem



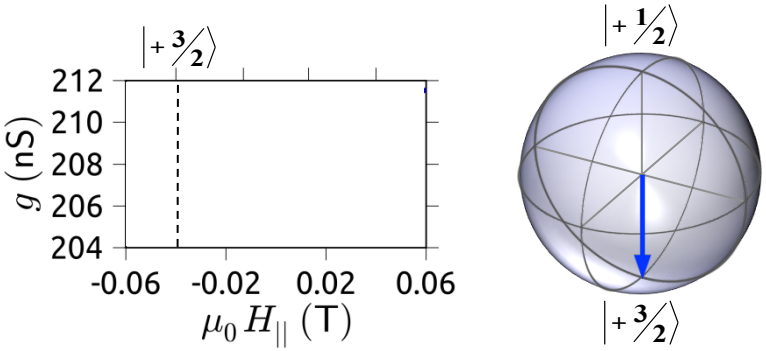
- applying a microwave pulse with f = 2.45 GHZ will rotate the vector

- different pulse lengths result in different rotation angles

Nuclear spin manipulation

I. Initialization:

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



Nuclear spin manipulation

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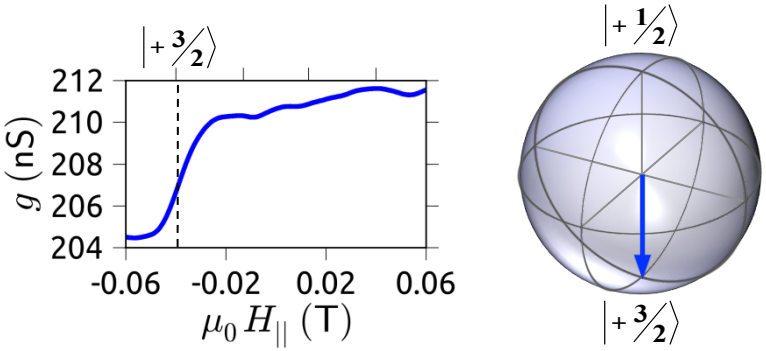
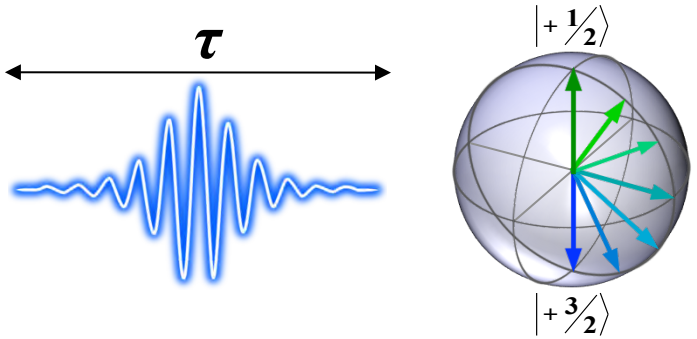


II. Pulse:
applying an oscillating electric field
with the frequency f and different
durations τ creates a coherent
superposition



Nuclear spin manipulation

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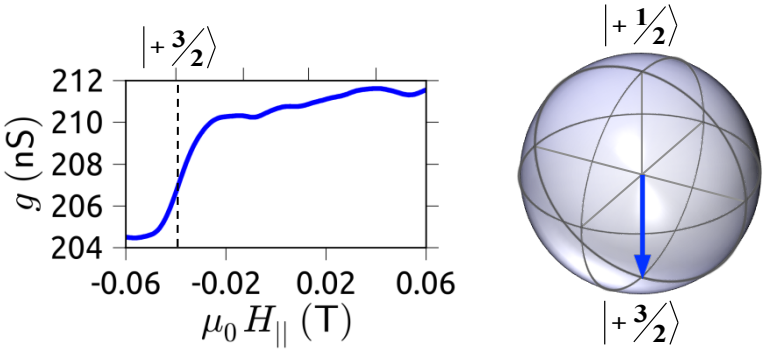

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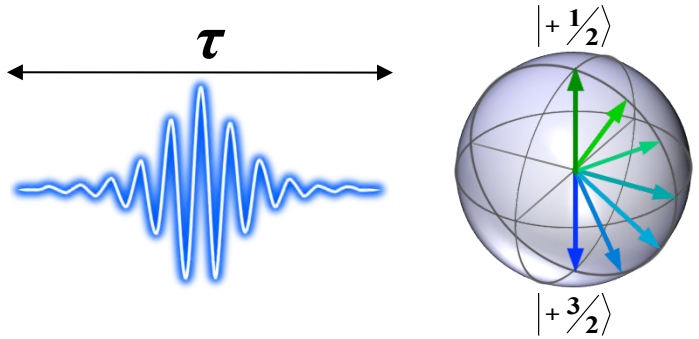
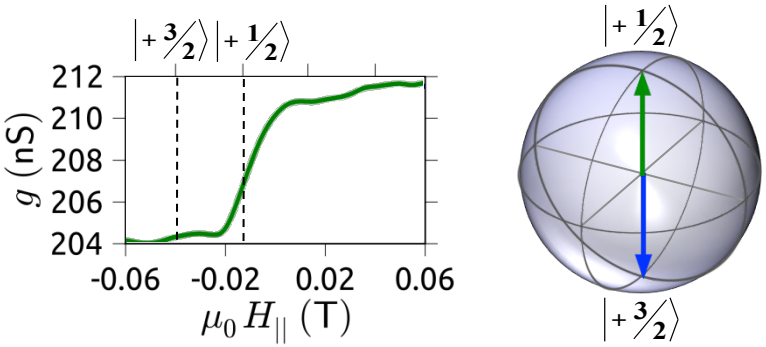


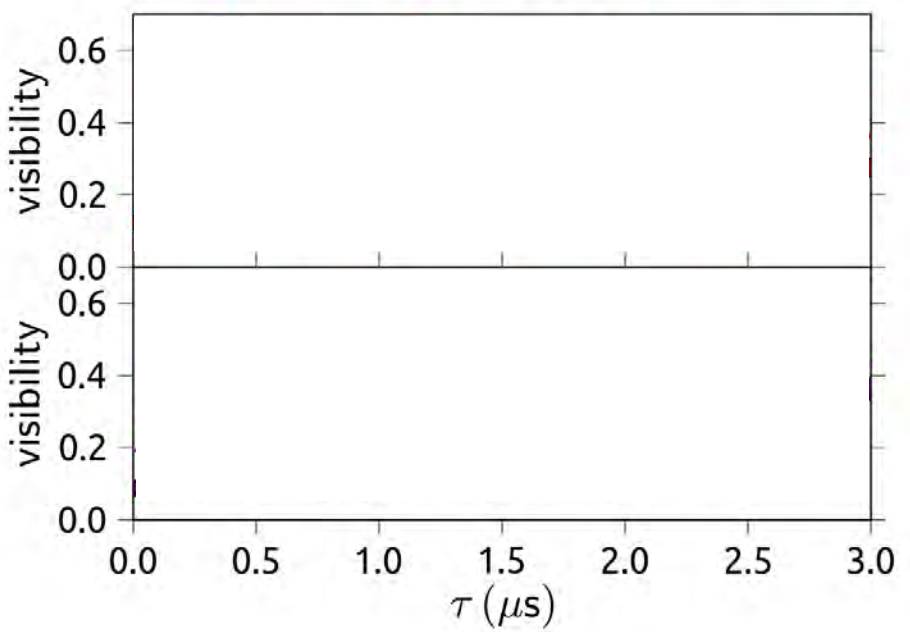
III. Read-out:

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



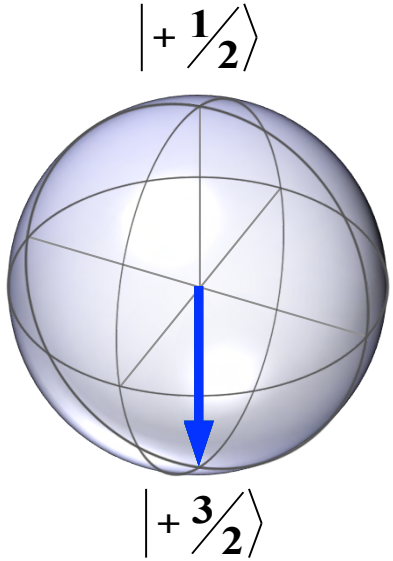
Nuclear spin manipulation

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



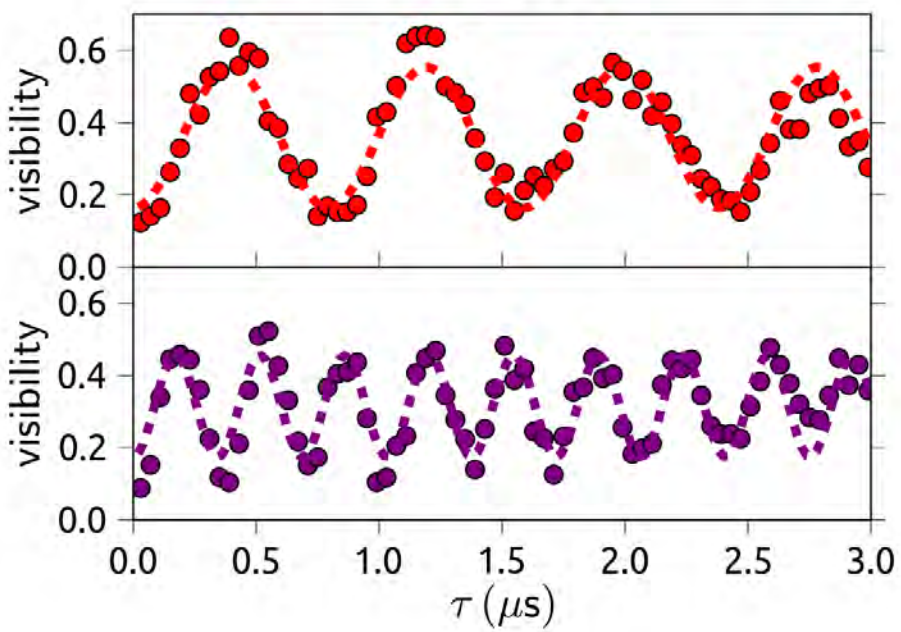
0 dBm

72 mT



Nuclear spin manipulation

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

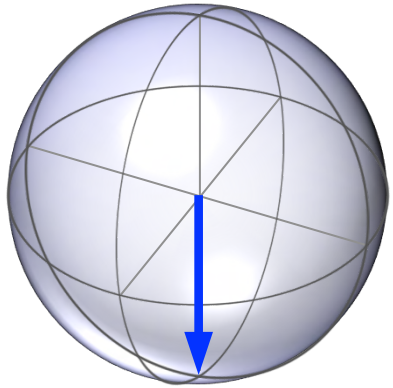


0 dBm

72 mT

2 dBm

170 mT



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

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

DC Stark effect

hyperfine Stark effct

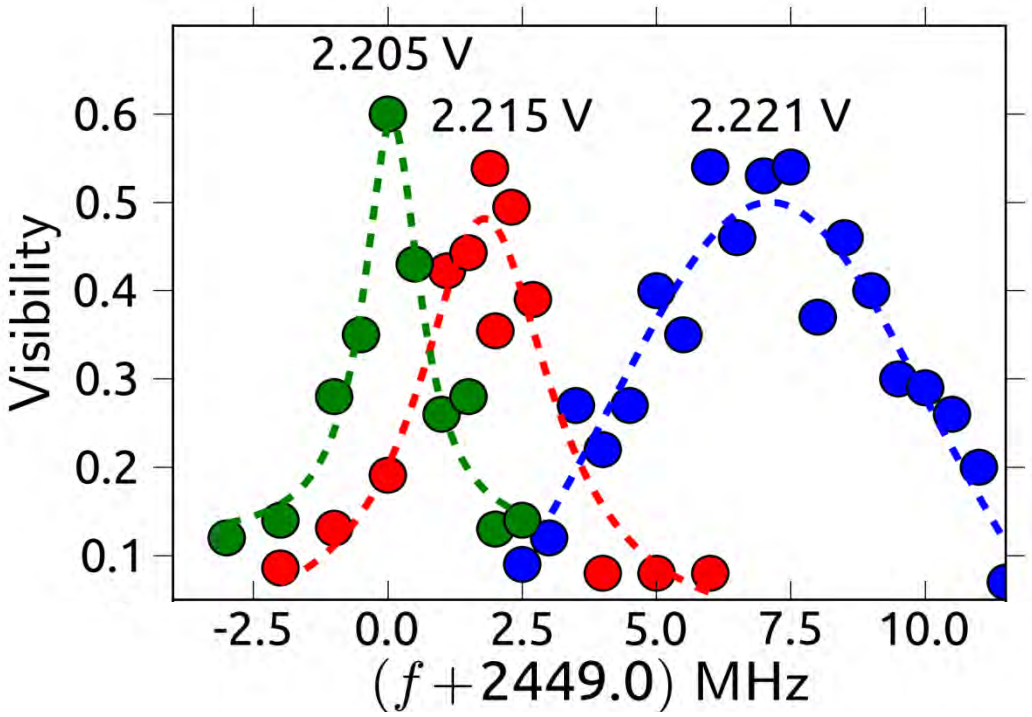
- AC electric field: **periodic** modulation of the hyperfine interaction
- DC electric field: **static** change of the hyperfine interaction

  → alter the level splitting (resonance frequency)

experiment

- Rabi amplitude largest at resonance → f_0
- V_g to change the static E-field
- Shift of f_0

electrical tunability of the nuclear qubit



Outline

Di Vincenzo 1996

Isolation

Read-out

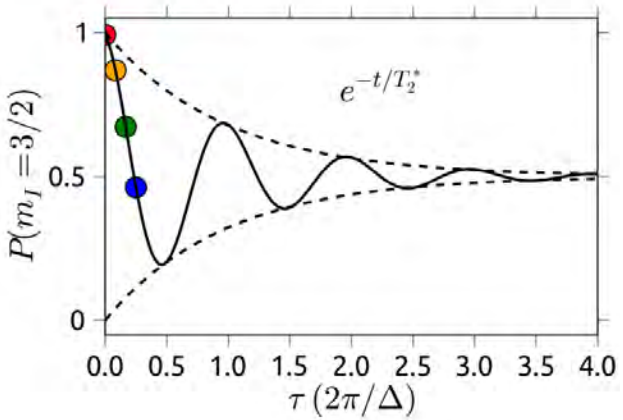
Initialization

Manipulation

Storage

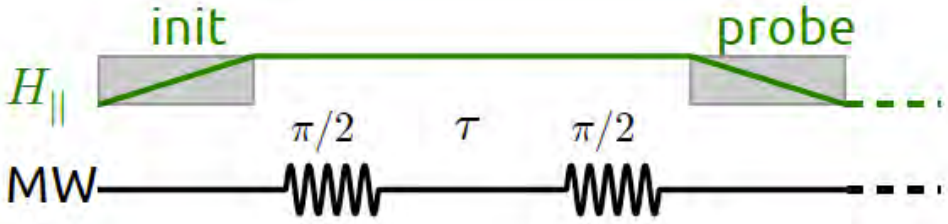
Grover's algorithm

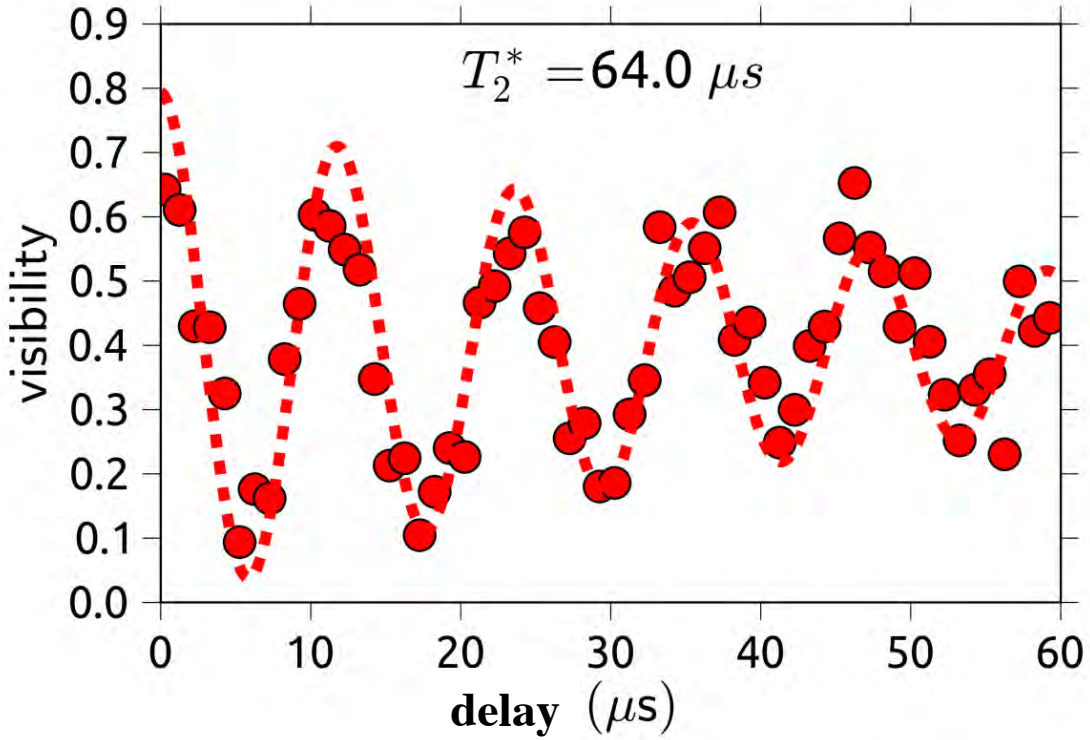
Basic quantum operation: Ramsey



- *Oscillation : Vector rotation*
- *Damping : Decoherence*

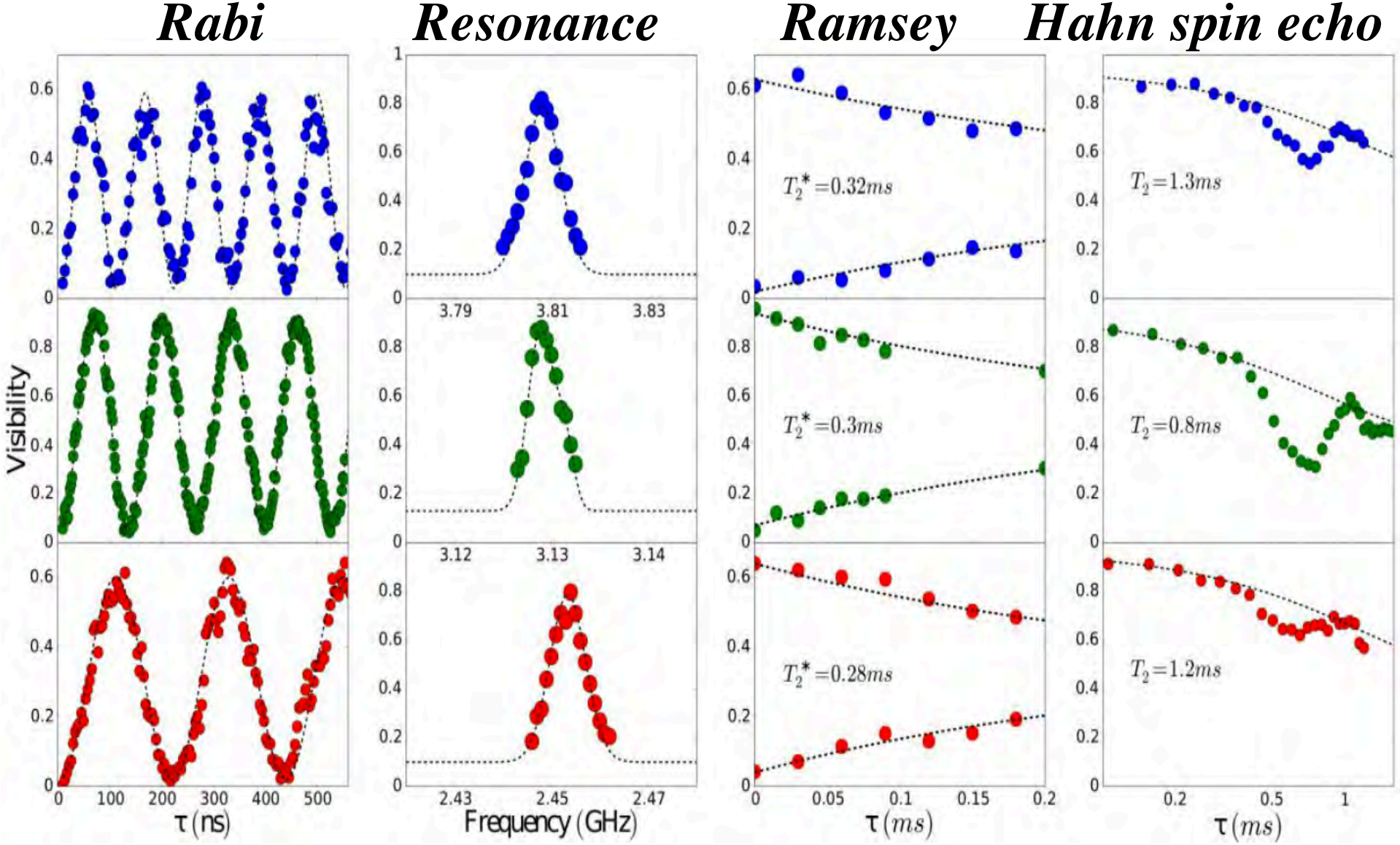
Basic quantum operation: Ramsey





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

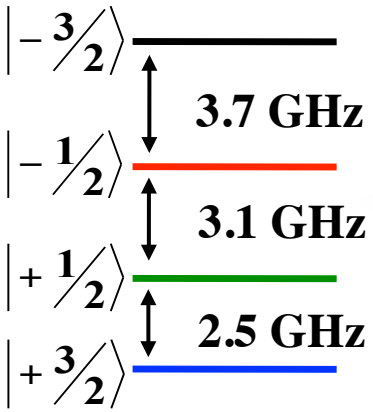
Basic quantum operation

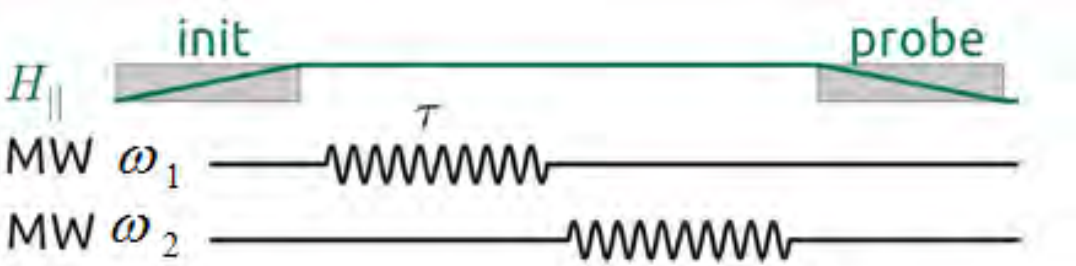


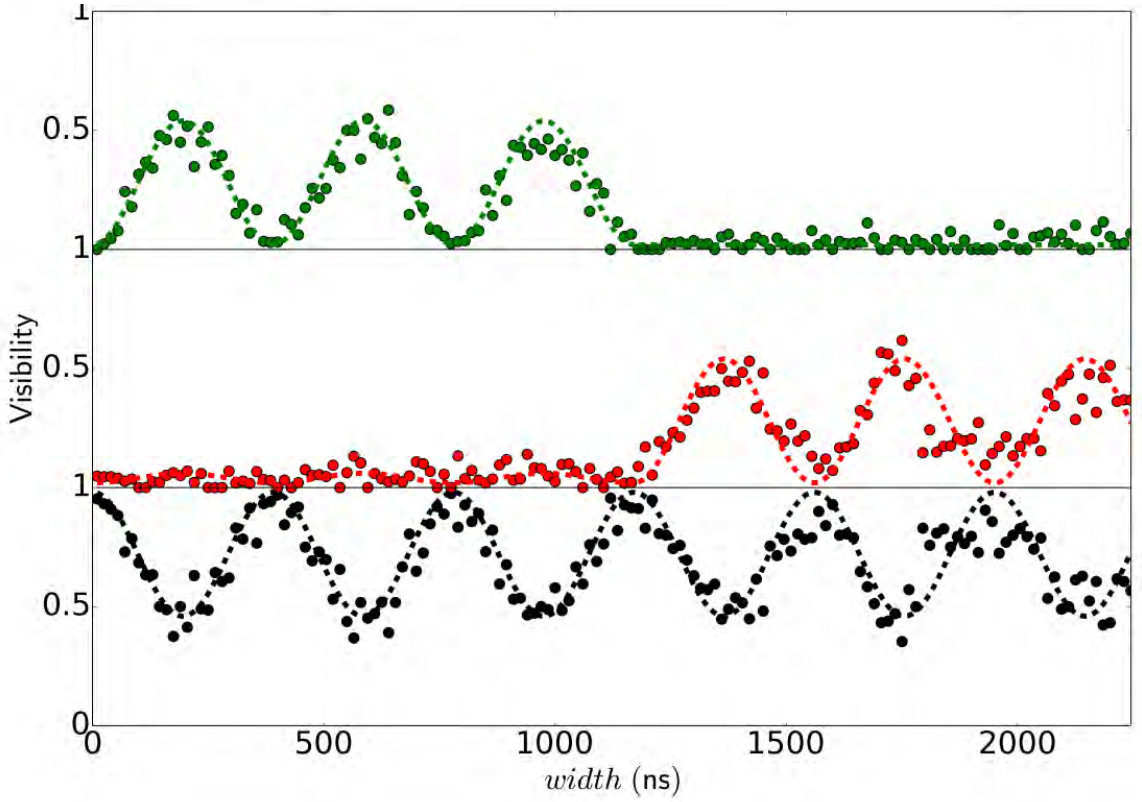
Multi-state manipulation – Quantum Gate

Coherent Pump probe

3 states

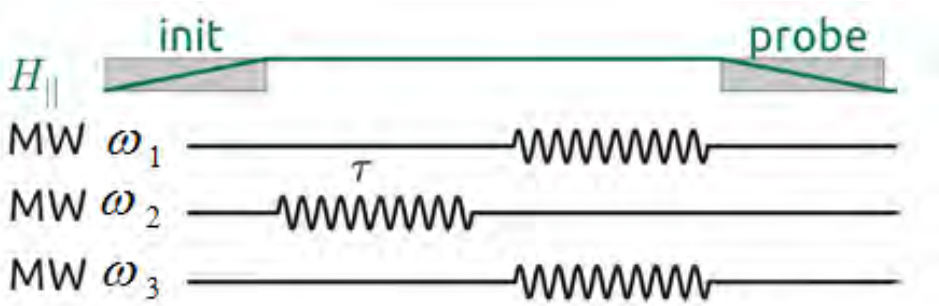




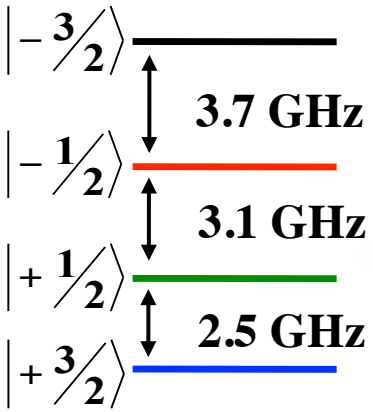


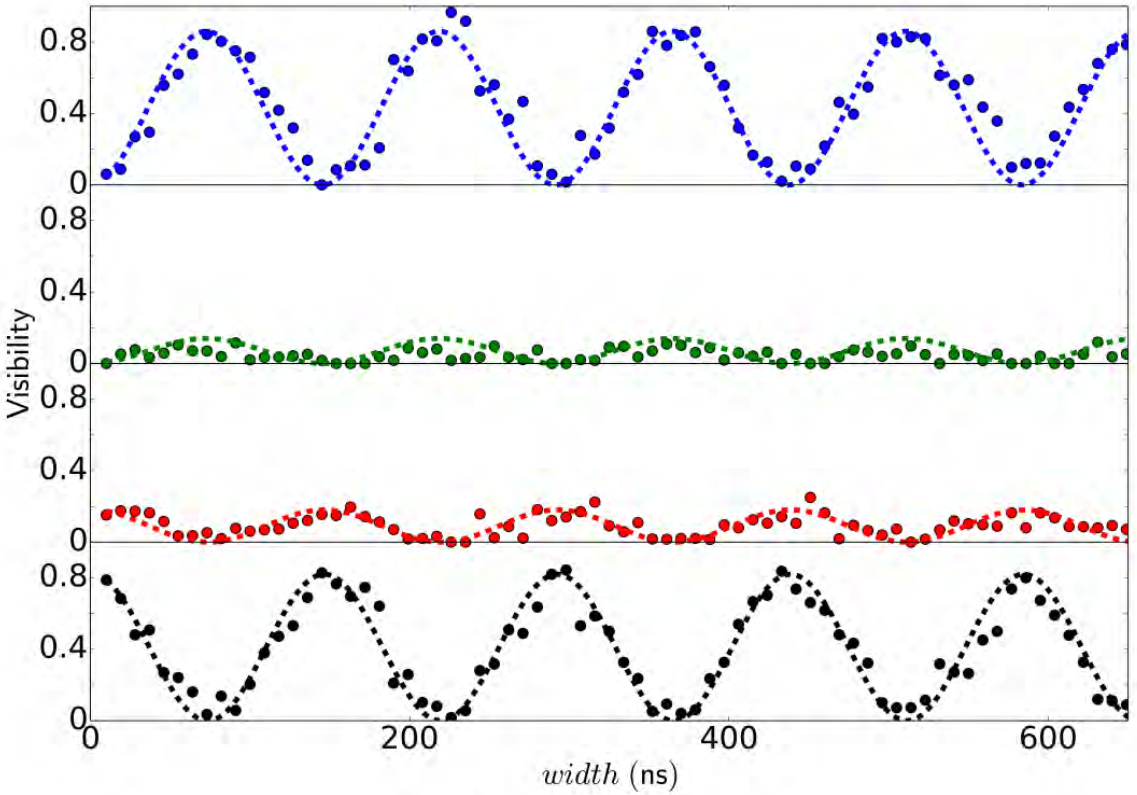
Multi-state manipulation – Quantum Gate

Coherent
Pump probe



4 states





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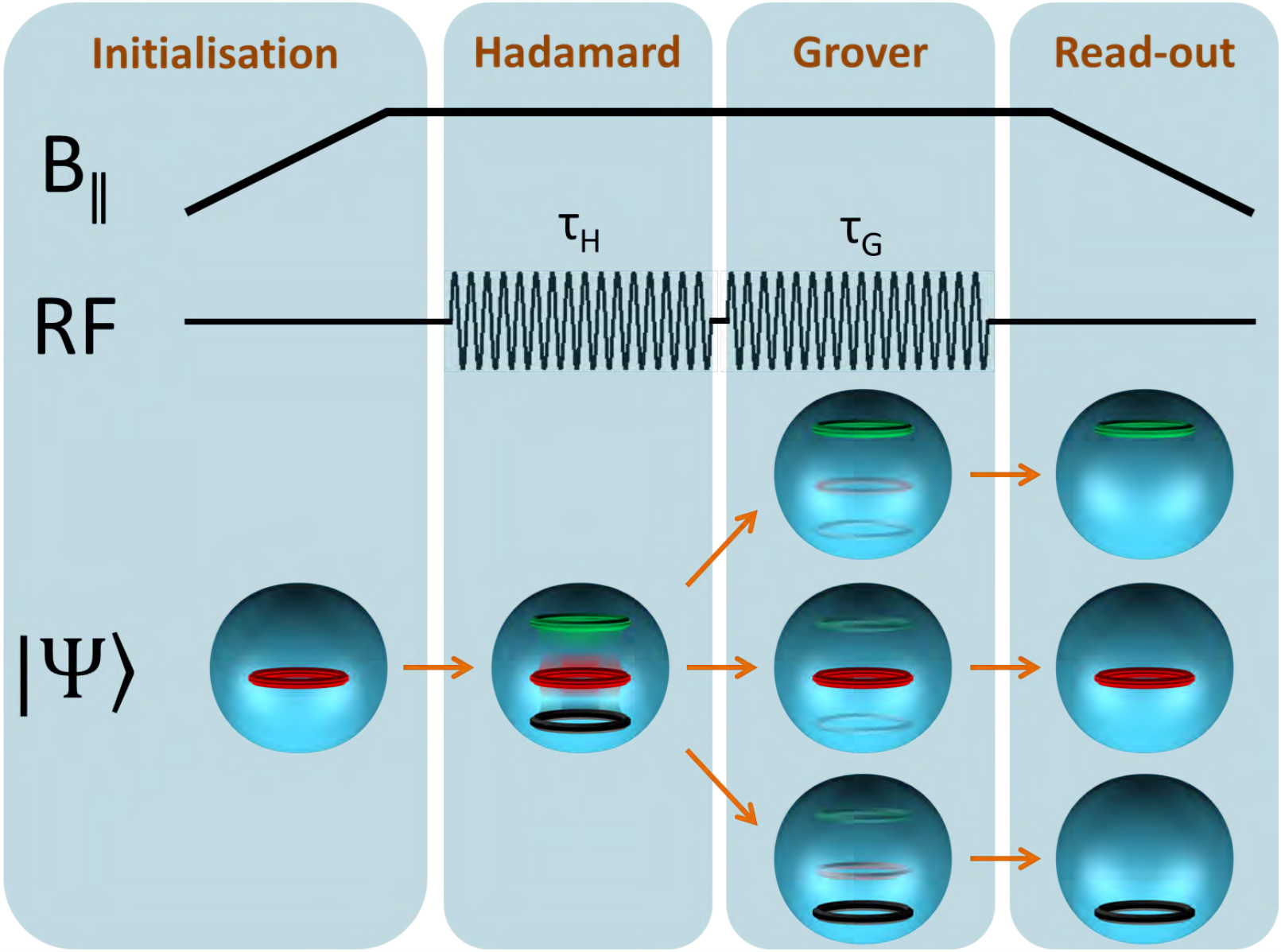
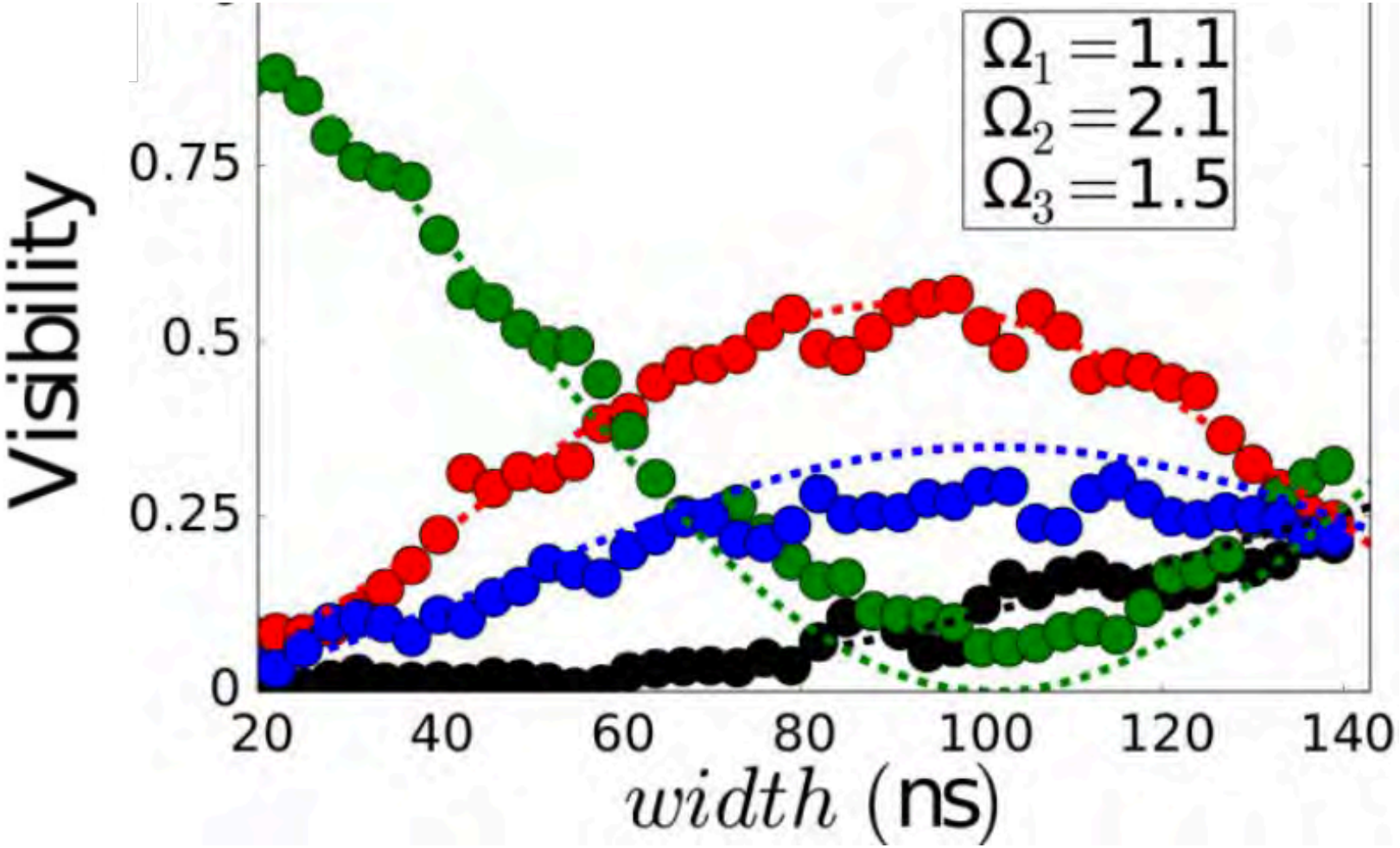


Grover's algorithm

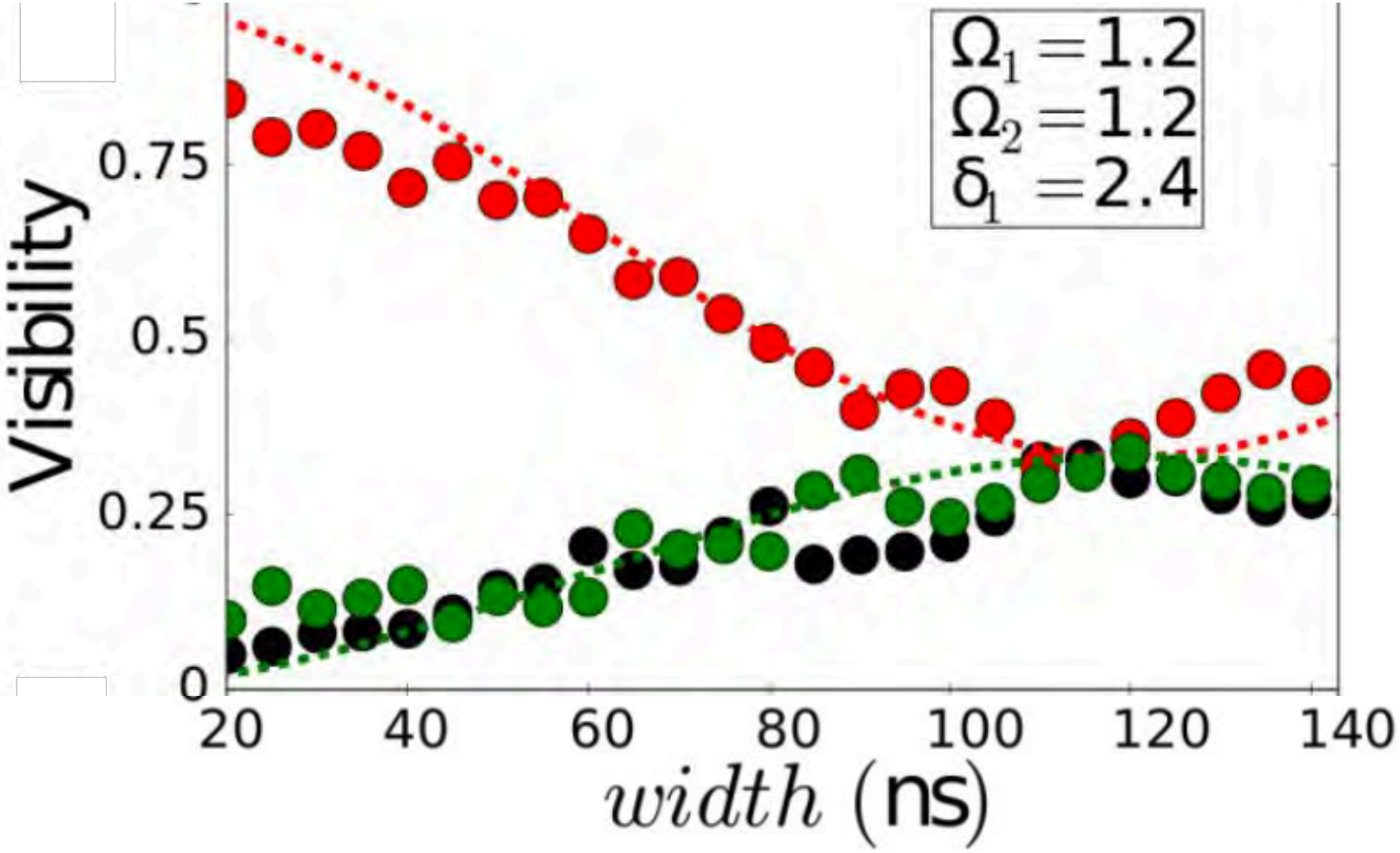
$$\left|+\tfrac{1}{2}\right\rangle \longrightarrow \frac{1}{\sqrt{4}}\left(\left|\tfrac{3}{2}\right\rangle+\left|\tfrac{1}{2}\right\rangle+\left|-\tfrac{1}{2}\right\rangle+\left|-\tfrac{3}{2}\right\rangle\right)$$

Hadamard gate with 4 states



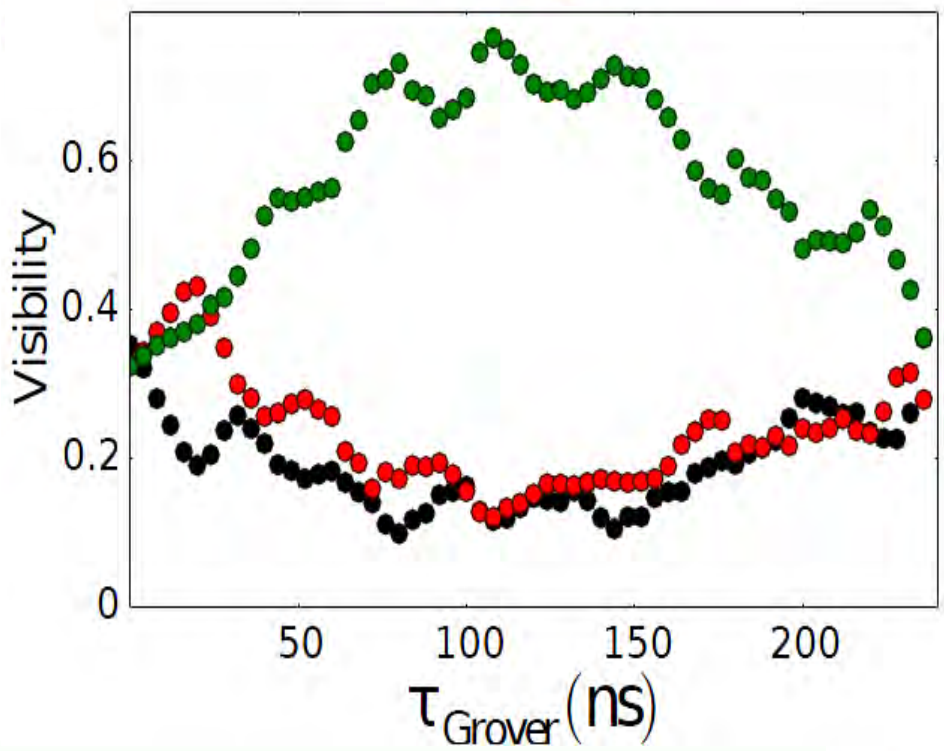
Grover's algorithm

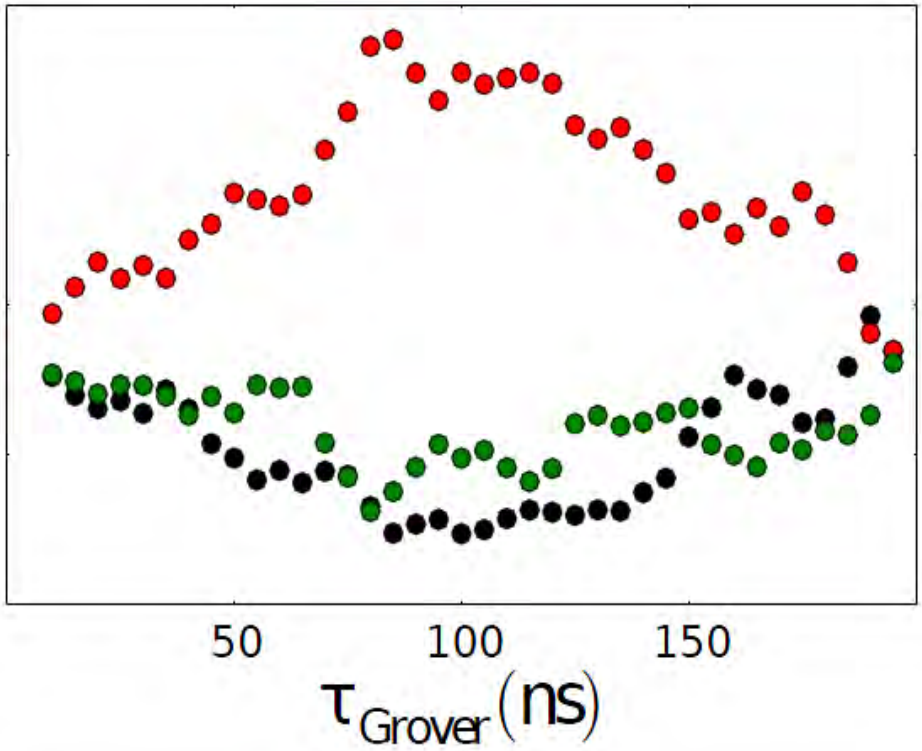
Hadamard gate with 3 states



Grover's algorithm

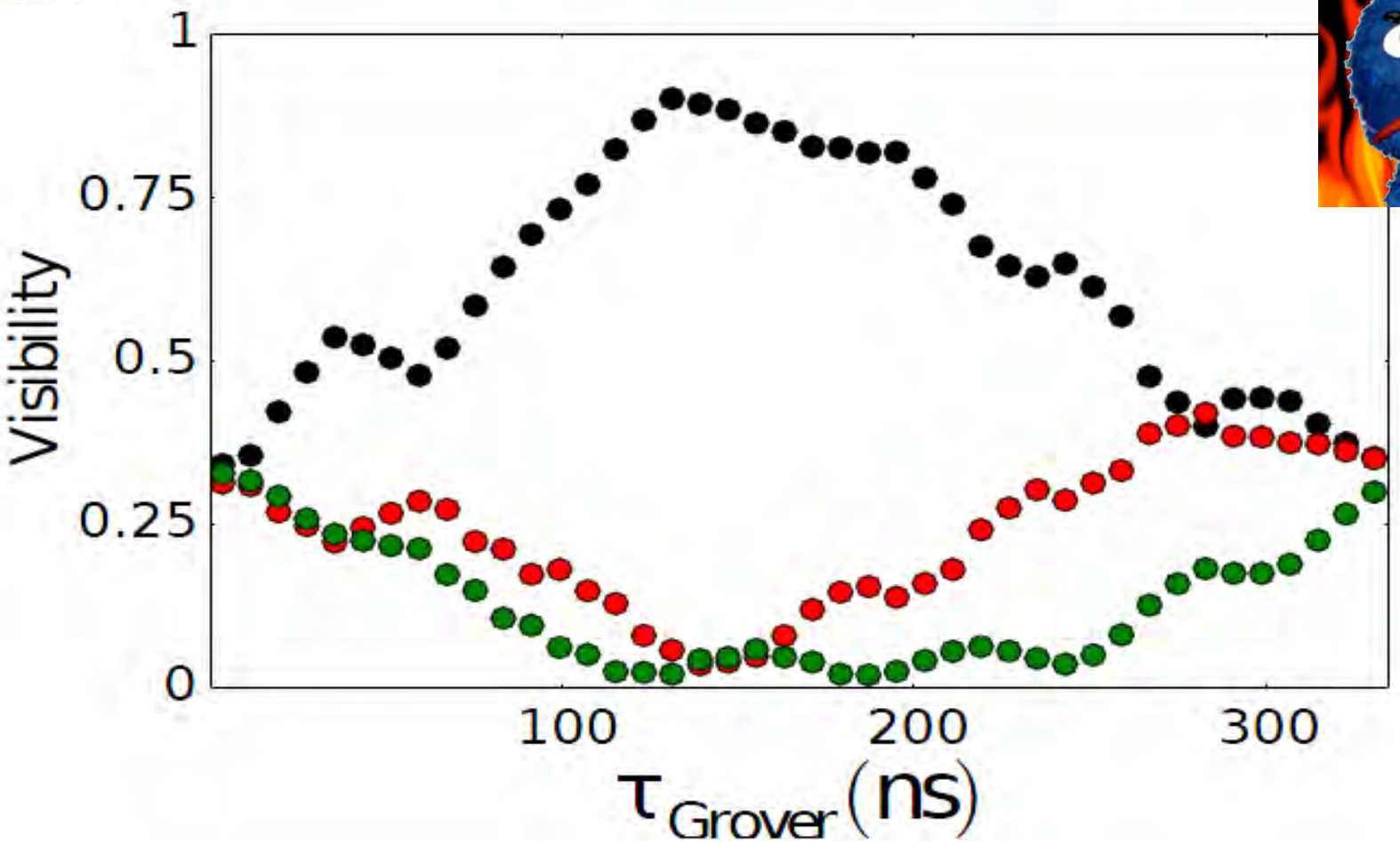
Selecting an element (state)





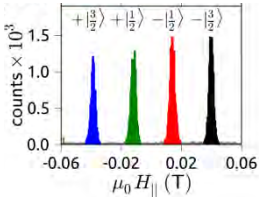
Grover algorithm

Selecting an element (state)

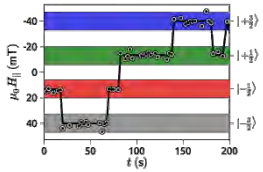


Single-molecule magnet qubits

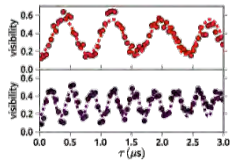
✓**isolation:** intrinsic to nuclear spins



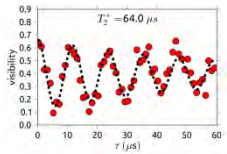
✓**read-out:** through the position of the conductance jumps



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



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

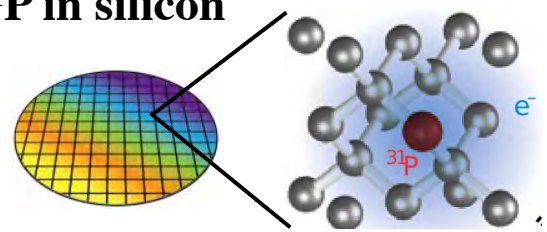


✓**information storage:** quantum information was stored for >300 μs

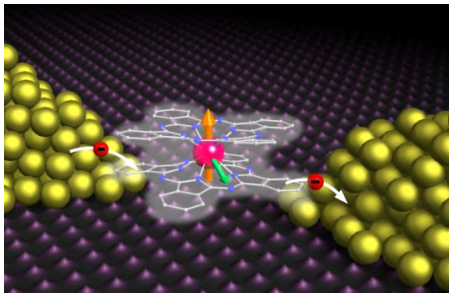
rover's algorithm

Quantum Technologies

^{31}P in silicon

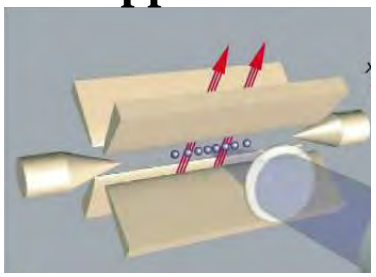


Nature 489, 541 (2012)



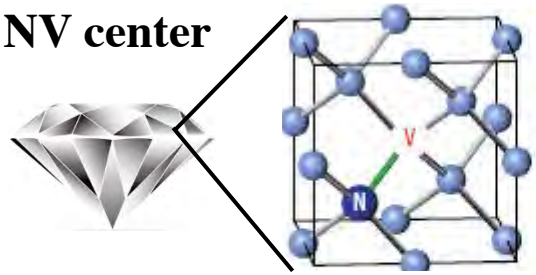
Nature 488, 357 (2012)
Science 344, 1135 (2014)

trapped ions

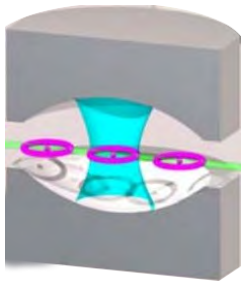


Nature 453, 1008 (2008)

NV center



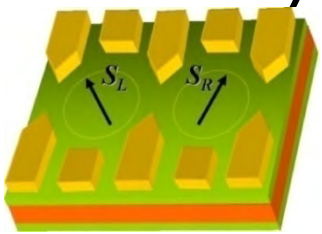
PRL 92, 076401 (2004)



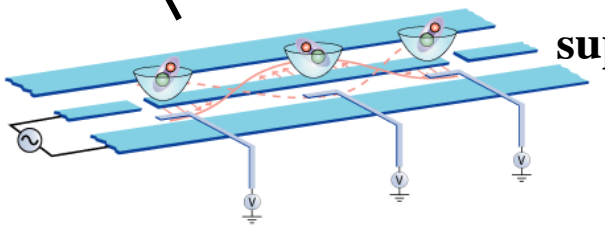
light in a cavity

Nature 446, 275 (2007)

quantum dots



Nature 491, 426 (2012) Nature 422, 766 (2006)



superconducting circuits

Nature 453, 1031 (2008)

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Staff

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E. Bonet
C. Thirion
W. Wernsdorfer

PhDs

M. Ganzhorn (2009-12): CNT res. + SMM
V. Nguyen (2009-12): CVD of CNT
M. Urdampilleta (2009-12): CNT + SMM
R. Vincent (2009-12): SMT + SMM
S. Thiele (2010-13): RF + SMT + SMM
C. Godfrin (2013-16): RF + SMT + SMM
J. Schönle (2014-17): CNT + SMM
S. Lumetti (2016-18): Graphene + SMM

Post-docs

J.-P. Cleuziou (2009-12): CNT + SMM
M. Lopes (2009-10): Raman + CNT
A. Candini (2009-10): Graphene + SMM
J. Järvinen (2009-12): RF + nanoSQUID+SMM
O. Geier (20012-14): CNT + SMM

Theoretical support

S. Florens, X. Blase, D. Feinberg …

Group of M. Affronte, Modena

Collaborations (Chemistry)

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Group of E. Brechin, Univ. of Edinburgh, UK
Group of R. Sessoli, D. Gatteschi, Univ. of Firenze, Italy
Group of A. Powell, Univ. Karlsruhe, Germany
Group of R. Clerac & C. Coulon, Univ. of Bordeaux
Group of D. Hendrickson, Dept. of Chemistry, San Diego
Group of T. Mallah, Univ. Paris Sud, Orsay
Group of M. Yamashita, Tokyo Univ.
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Group of A. Cornia, Univ. of Modena, Italy
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Group of V. Marvaud & M. Verdaguer
Univ. P. et M. Curie, Paris
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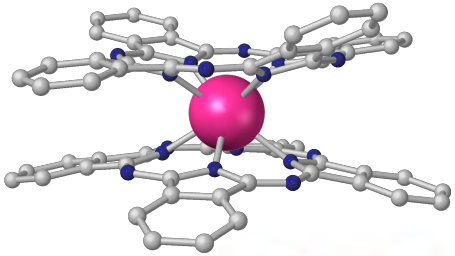
Group of K. Dunbar, Univ. of Texas, US
Group of M. Julve, Univ. of Valence, Spain
Group of P. Mialane, Univ. of Versailles, France
Group of E. Coronado, Univ. of Valence, Spain
Group of M. Ruben, FZ Karlsruhe, Germany
and many more …



Support

(2008-2013)

(2008-2016)

(2013-2016)

(2008-2013)

Thank you !!!