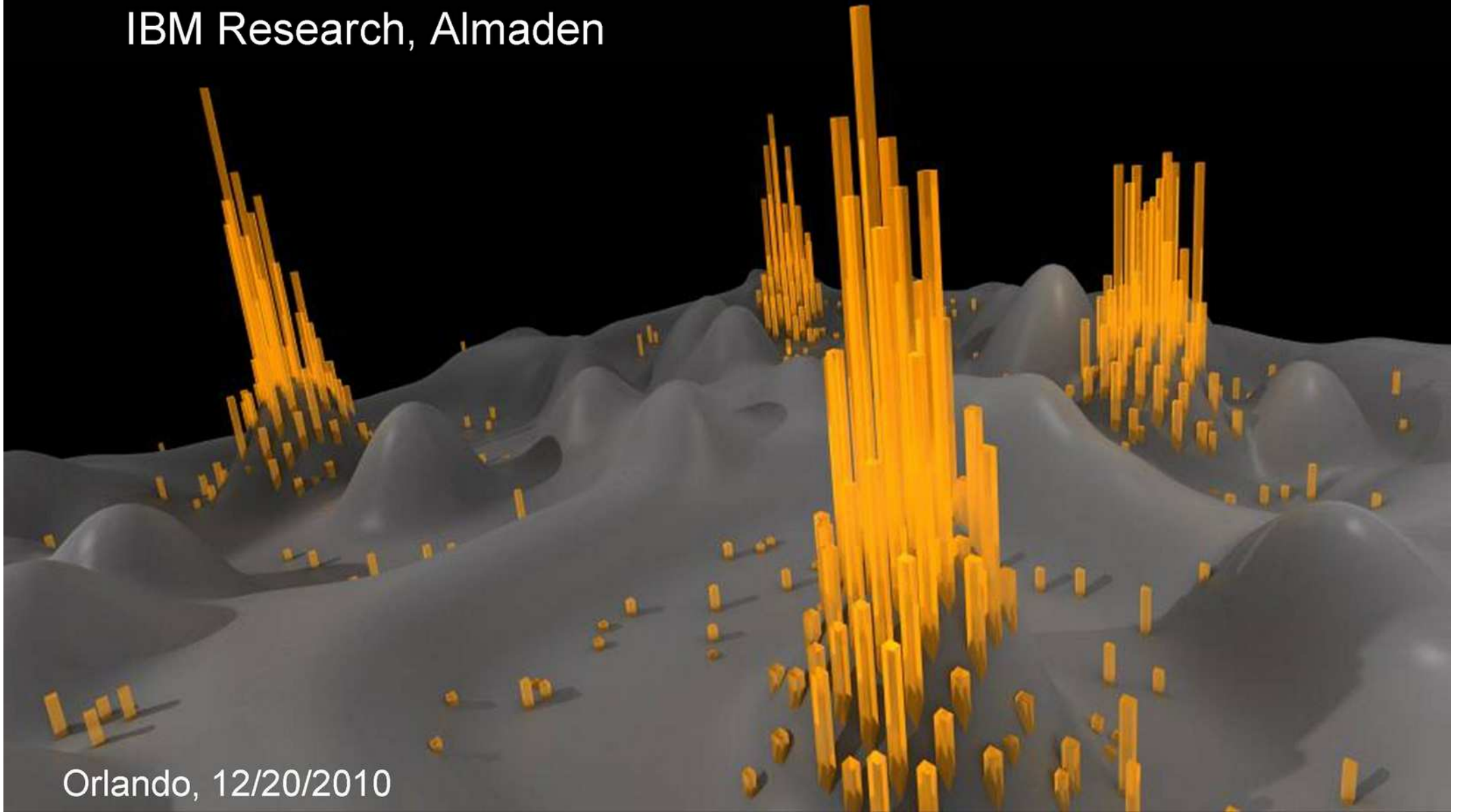


Quantum Tunneling of Magnetization in individual atoms

Sebastian Loth

IBM Research, Almaden



Orlando, 12/20/2010

ARC



Andreas
Heinrich



Don
Eigler



Chris
Lutz



Bruce
Melior



Susanne
Baumann



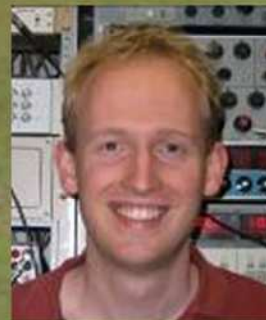
Markus
Etzkorn



Kirsten v.
Bergmann



Markus
Ternes



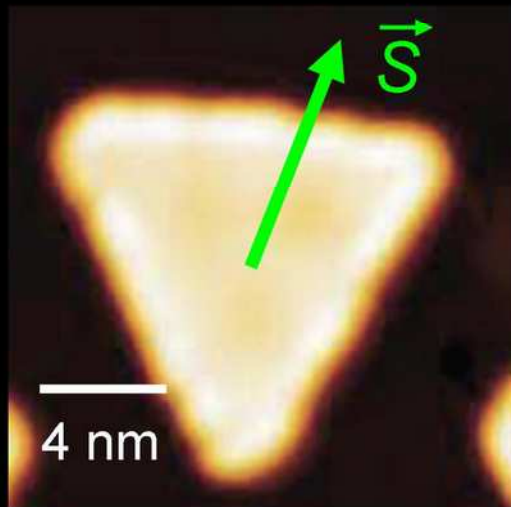
Sander
Otte



Cyrus
Hirjibehedin



Classical Magnets



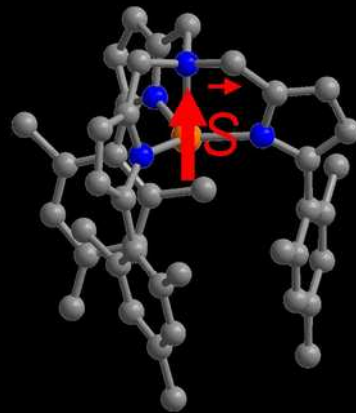
Co island on Cu(111)

~ 3000 atoms
 $S > 1000$

Classical Magnetization

Oka *et al.* Science (2010)

Quantum Magnets



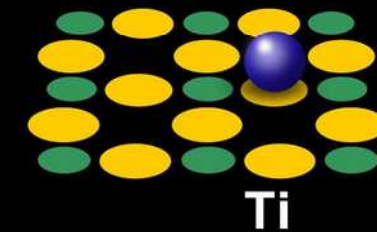
$[(\text{tpa}^{\text{Mes}})\text{Fe}]^-$ molecule

1 magnetic atom
 $S \approx 2$

Magnetic Tunneling

Freedman *et al.* JACS (2010)

Spins



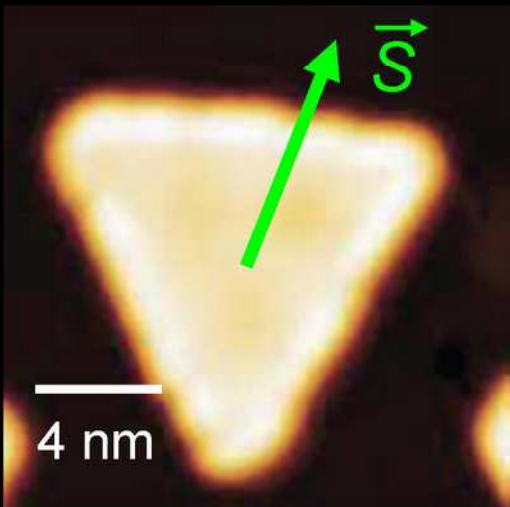
Ti atoms on Cu_2N / Cu(100)

1 atom
 $S = 1/2$

Kondo effect

Nature Physics (2008)

Magnetism in STM: Magnetic Islands

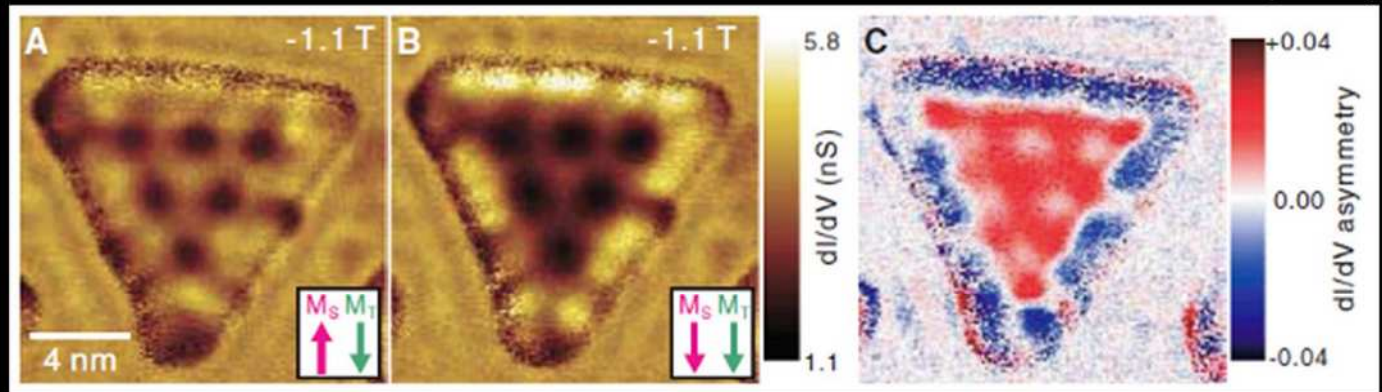


Oka *et al.* Science (2010)

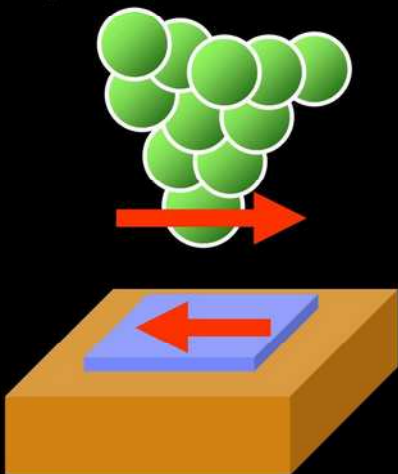
➤ Spin-polarized STM tips resolve the magnetic orientation

- ❖ Single domain magnets: One Macro-'Spin'

Oka *et al.* Science (2010)

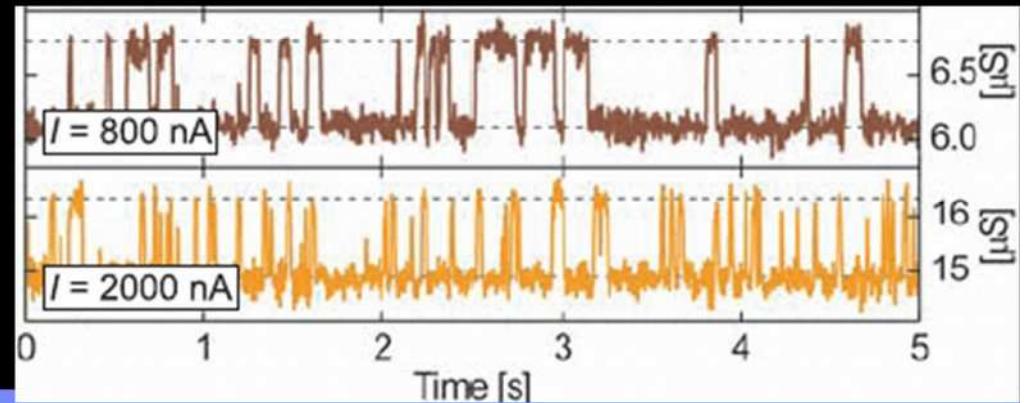


Spin-polarized tip



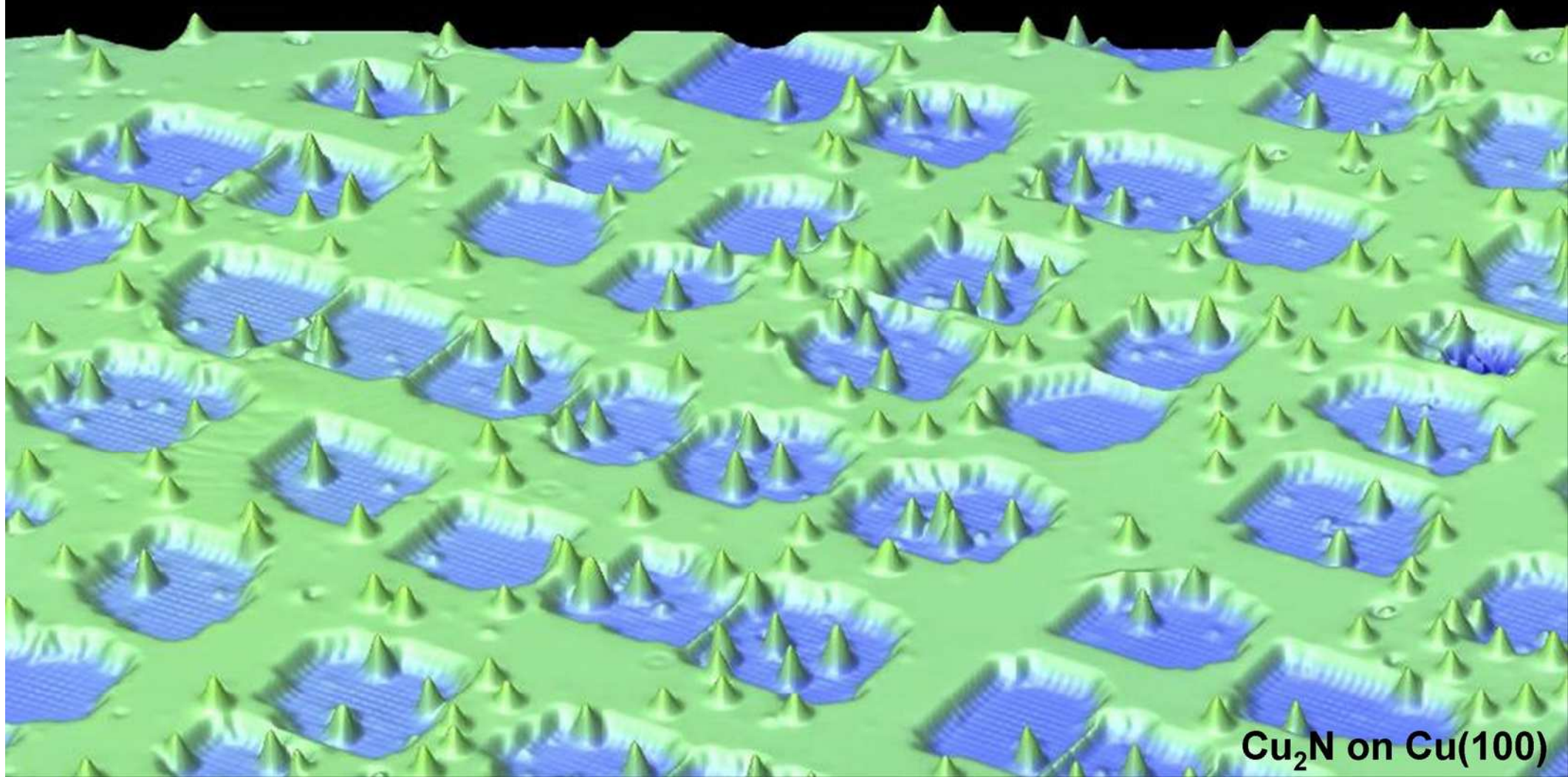
- ❖ Thermally-induced or Spin-torque assisted switching observable.

Krause *et al.* Science (2007)



Magnetism in STM: Individual atoms

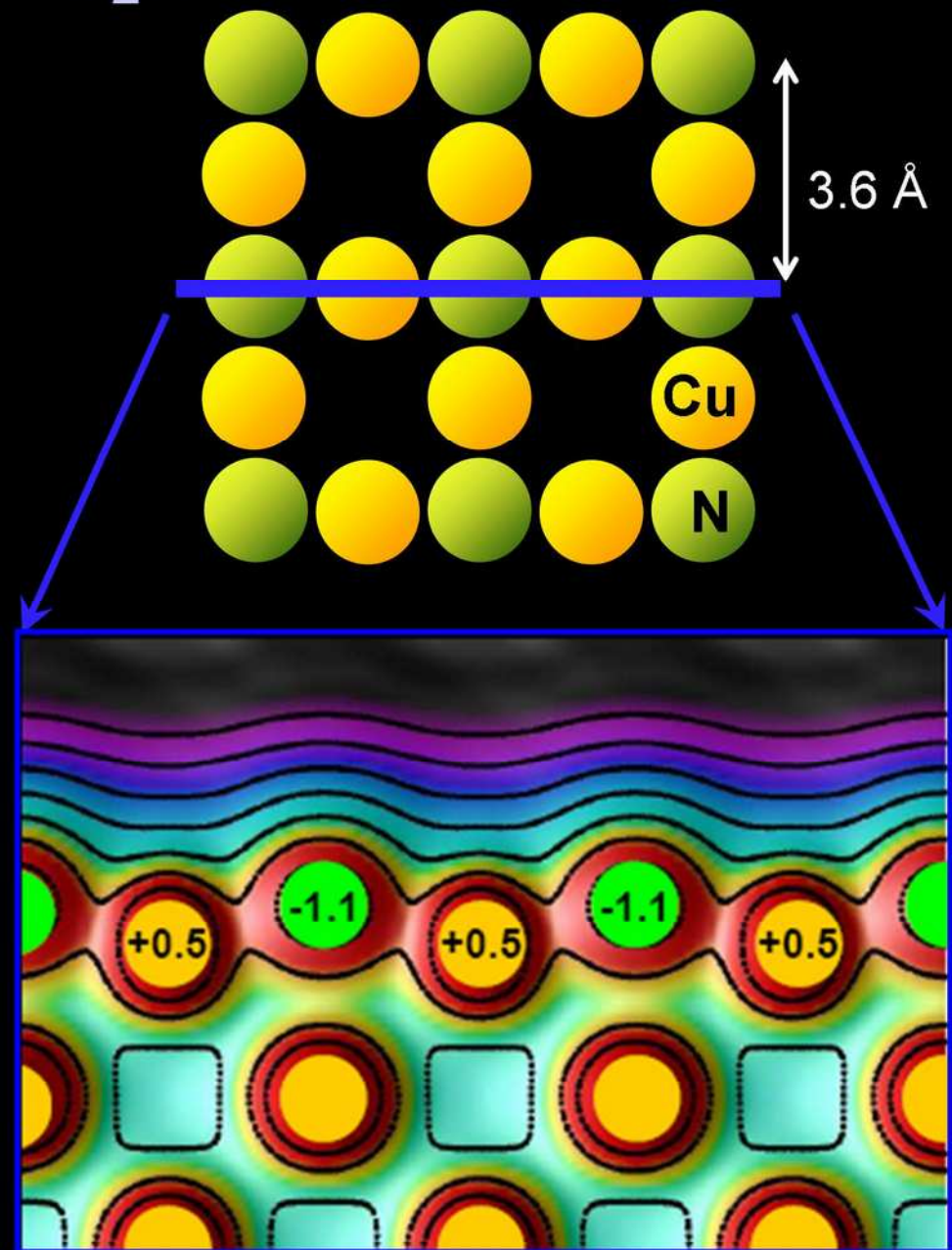
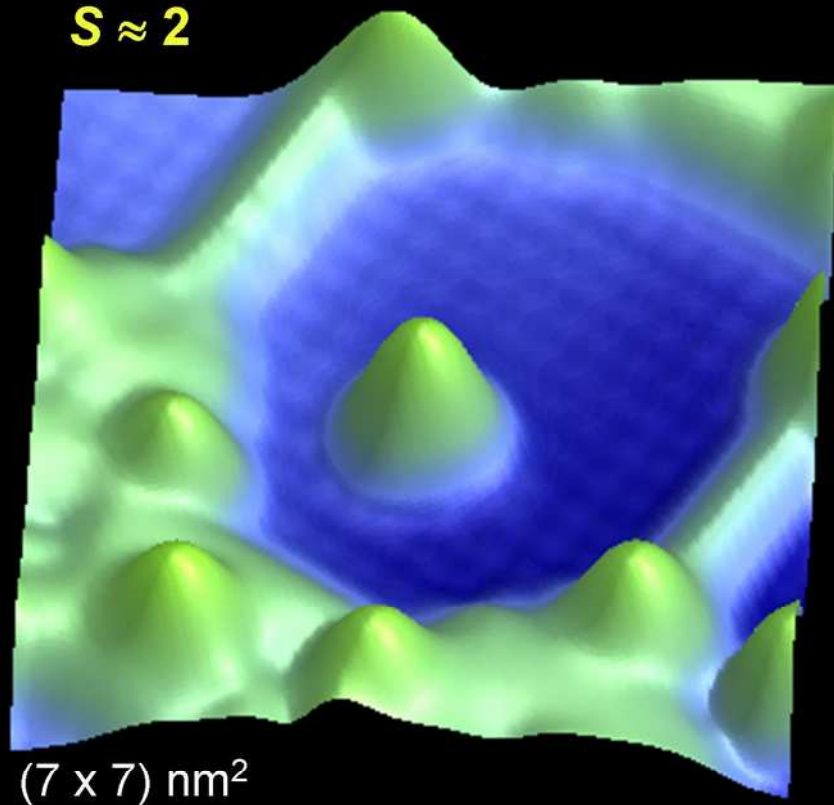
- Transition metal atoms (Fe, Mn, Cu)
- Substrate Cu_2N on $\text{Cu}(100)$
- Quantum mechanical description of spins necessary



Cu_2N on $\text{Cu}(100)$

Transition metal atoms on Cu_2N

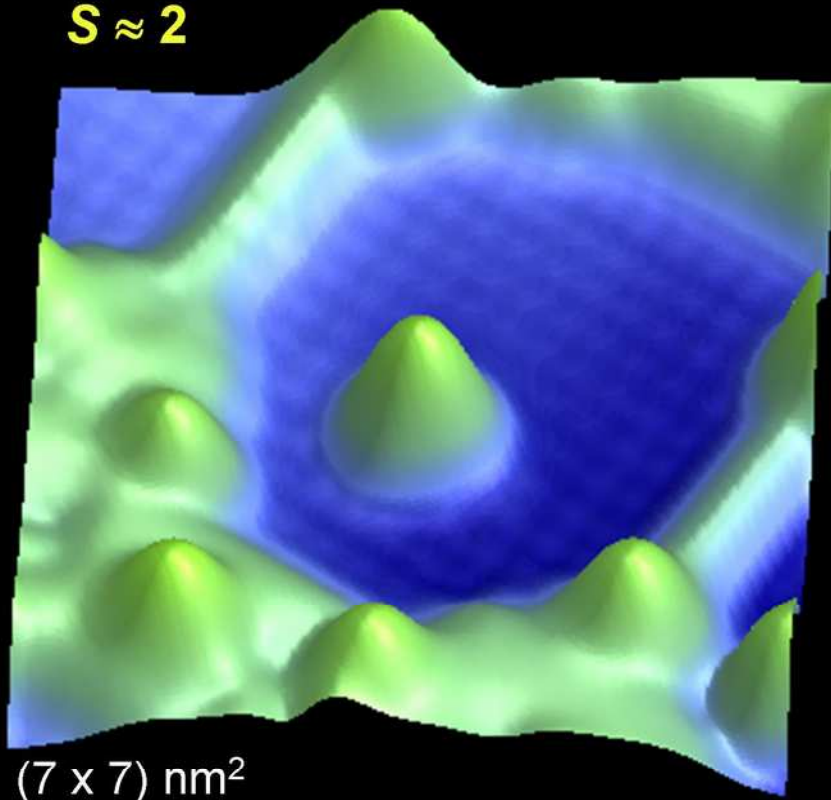
Mn - Cu_2N
 1 magnetic atom
 $S \approx 2$



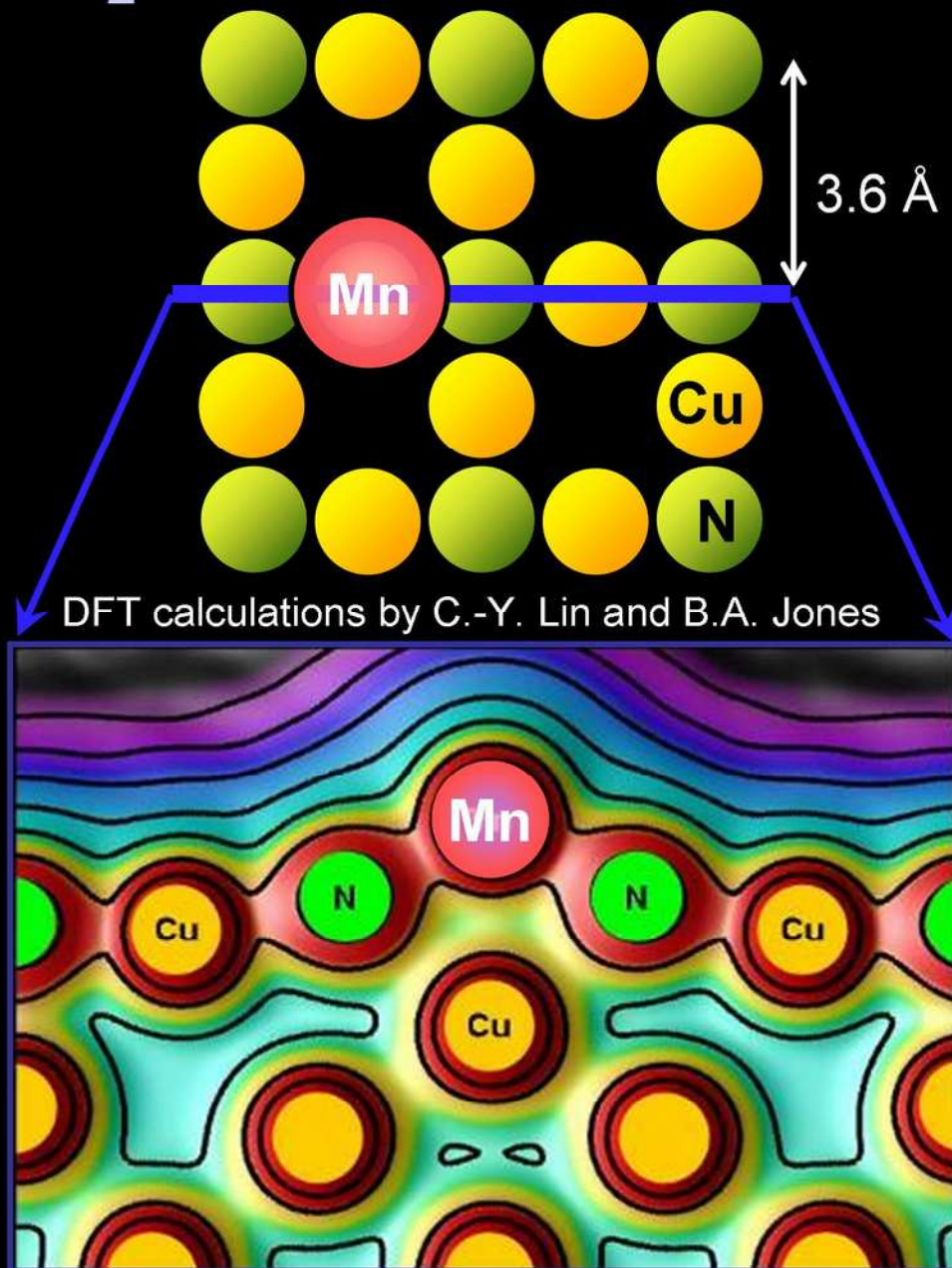
➤ **N forms planar molecular network**

Transition metal atoms on Cu_2N

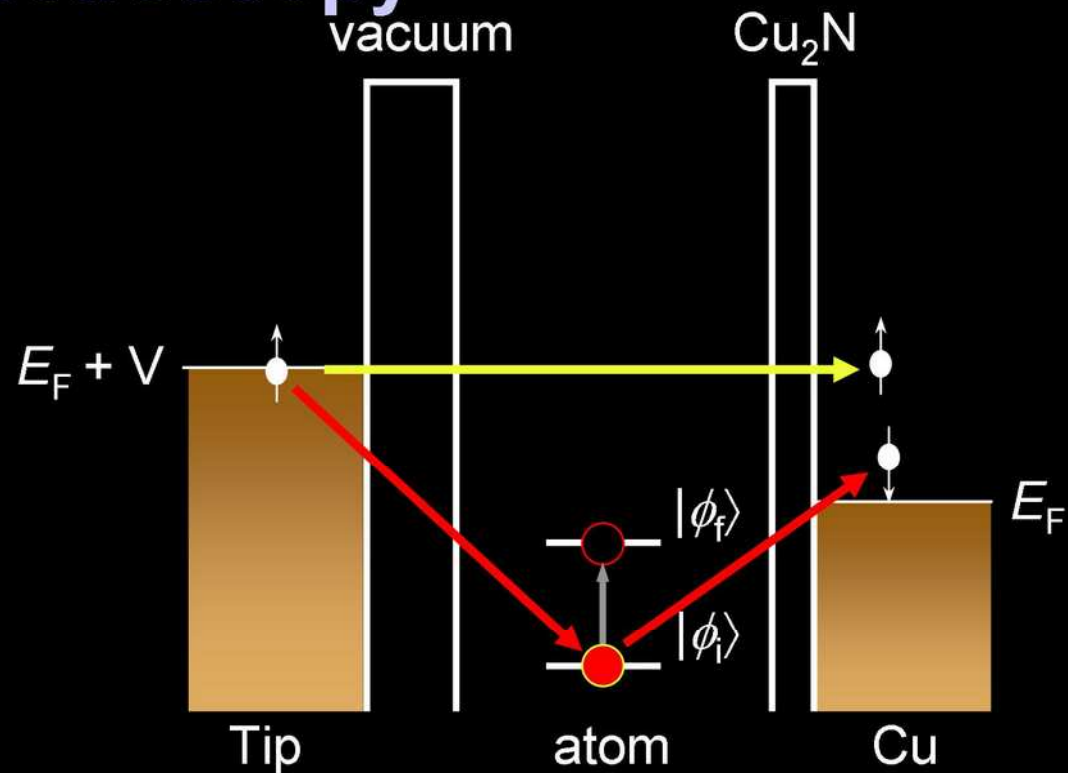
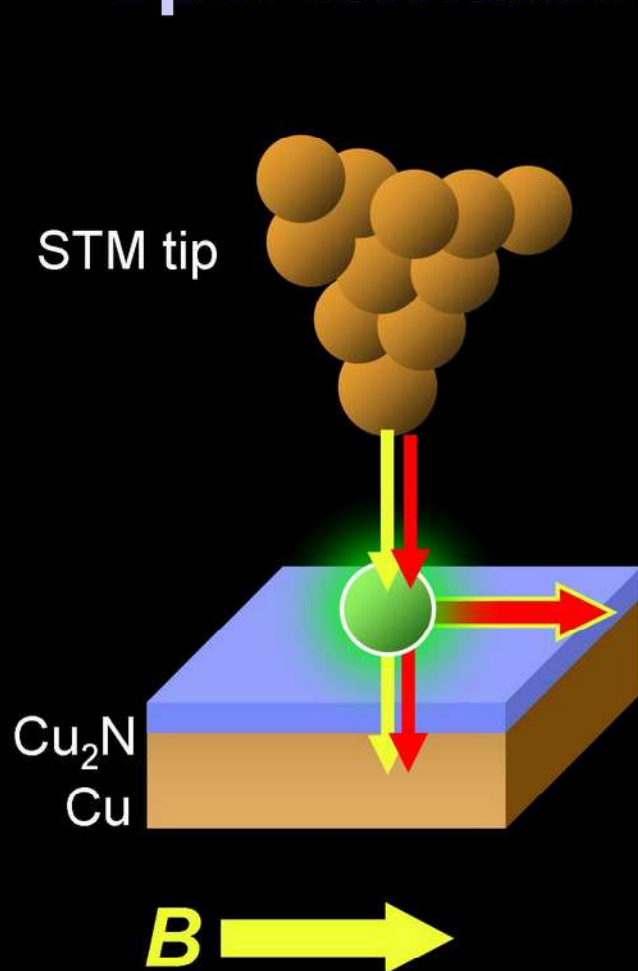
Mn - Cu_2N
 1 magnetic atom
 $S \approx 2$



- **N forms planar molecular network**
- **Mn is held in this network**



Spin excitation spectroscopy

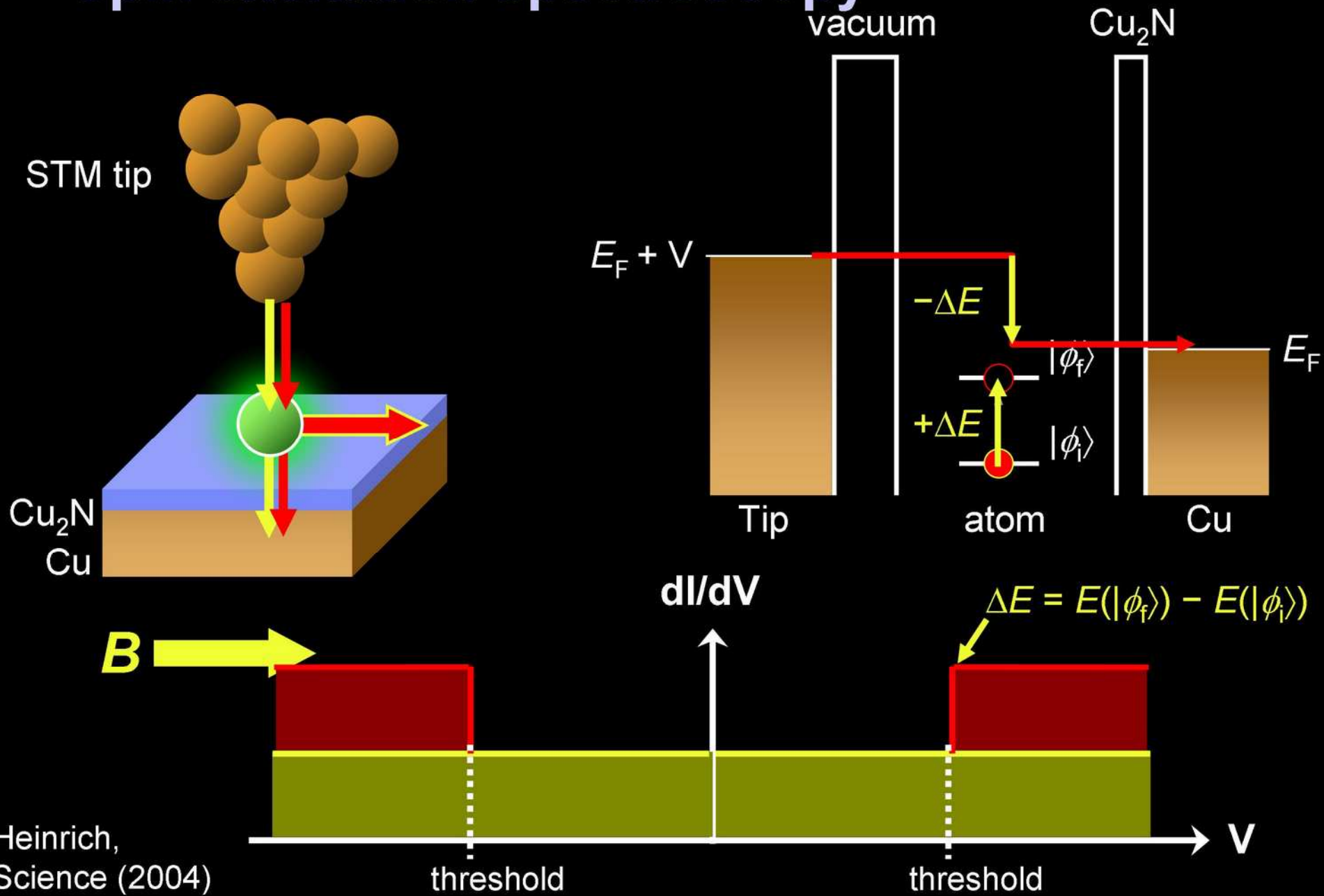


Inelastic co-tunneling
Interaction Hamiltonian:

$$\vec{s} \cdot \vec{S} + u$$

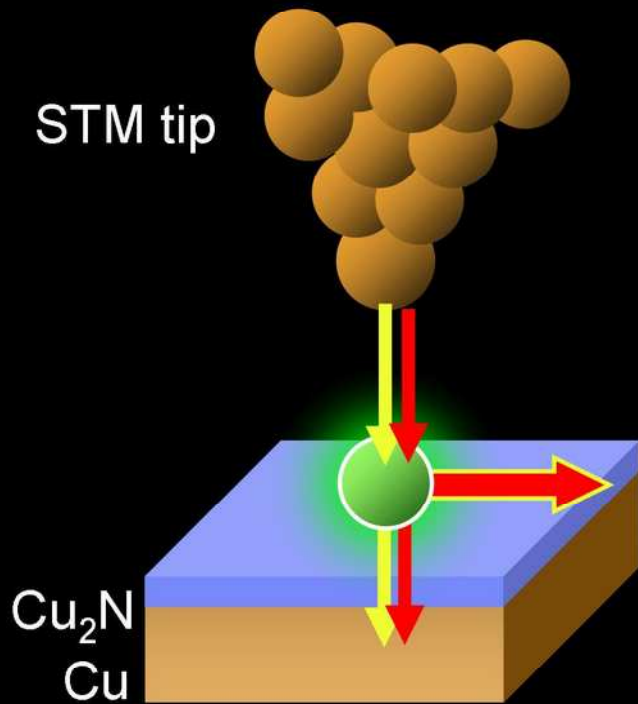
e.g. Schrieffer, Wolff Phys. Rev. **149** 491 (1966)

Spin excitation spectroscopy

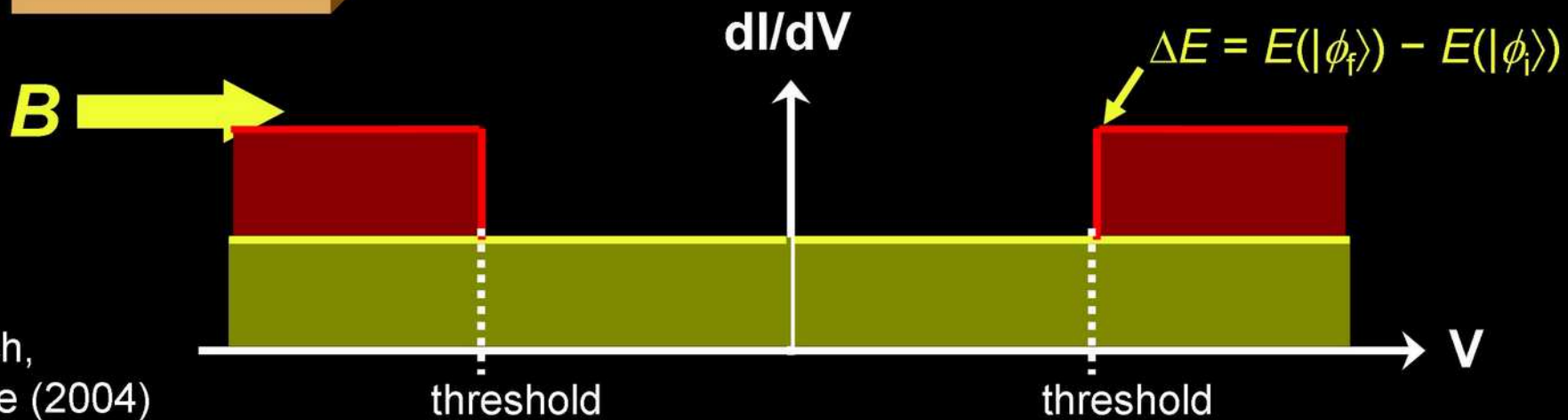
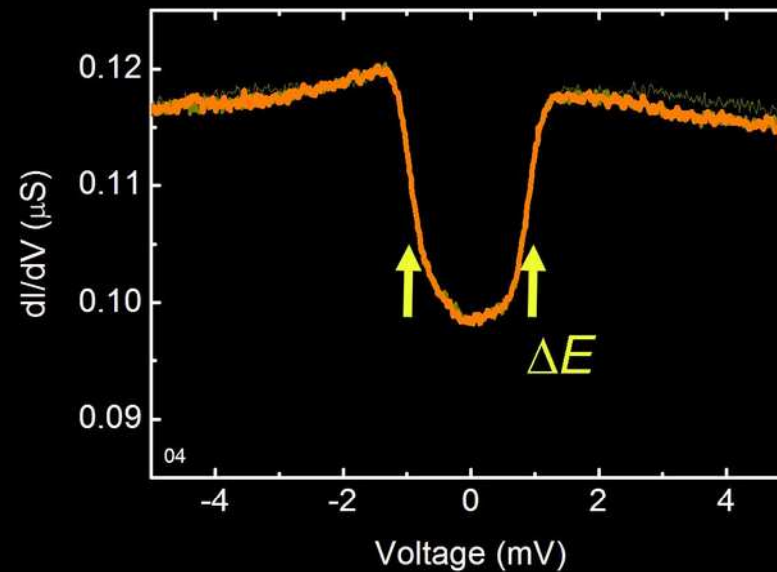


Heinrich,
Science (2004)

Spin excitation spectroscopy



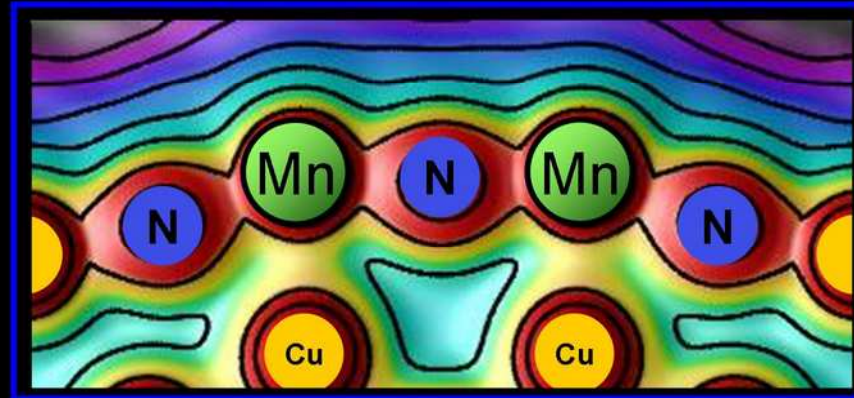
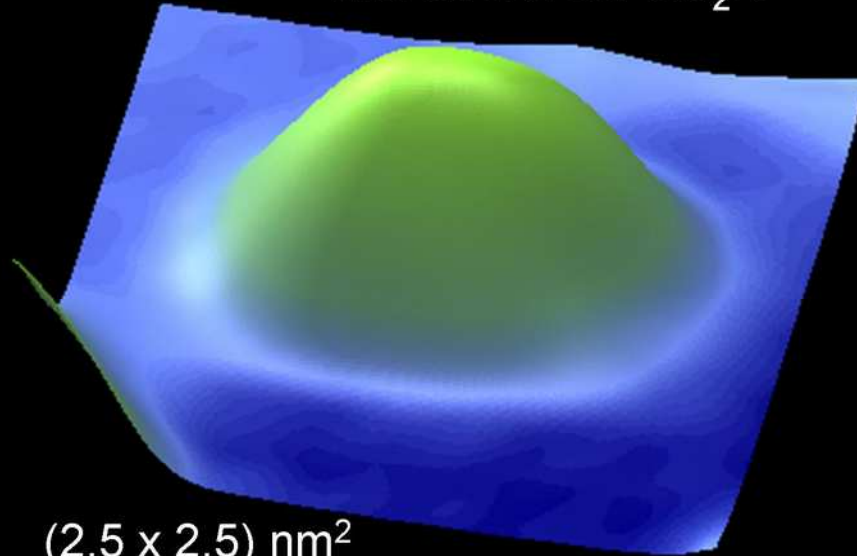
Mn atom



Heinrich,
Science (2004)

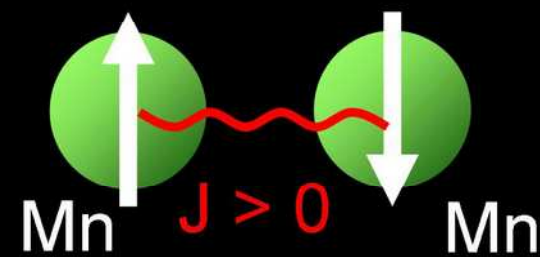
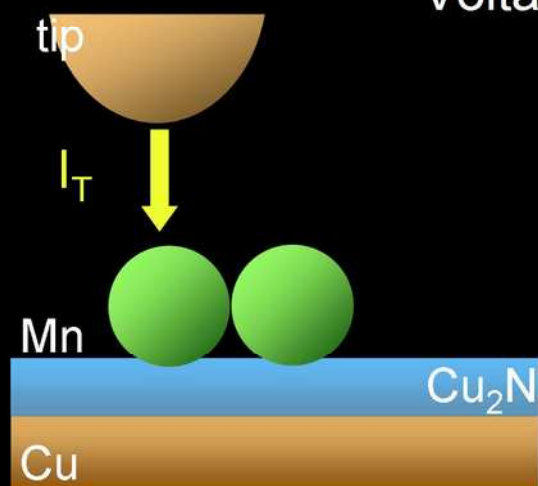
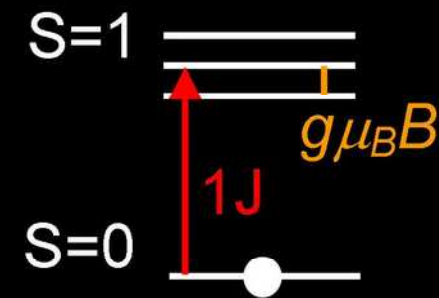
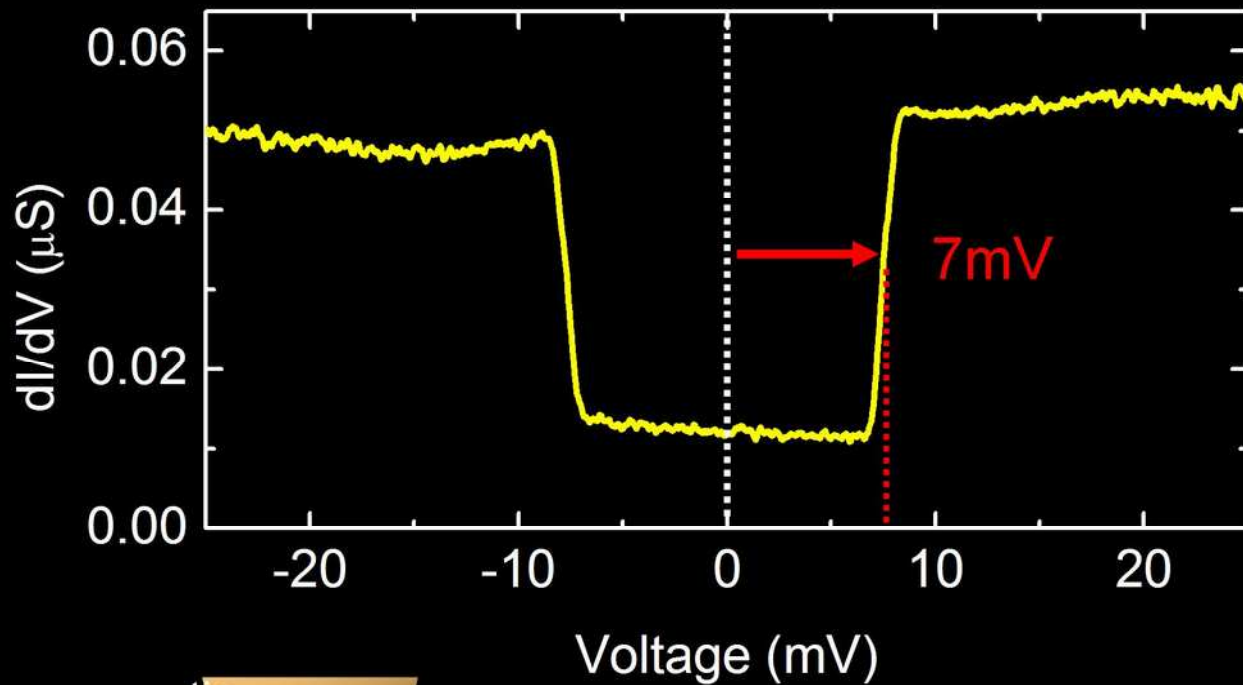
Spin excitations in Mn-N-Mn dimer

Mn dimer on Cu_2N



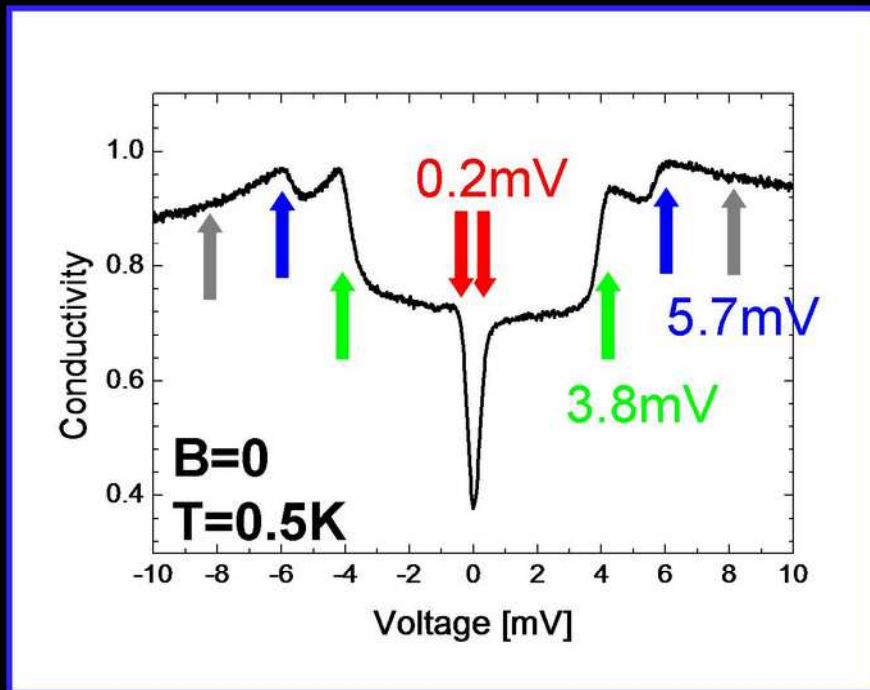
- **Polar covalent bonds between Mn and N on Cu_2N**
- **surface-embedded molecular magnet (?)**

Spin excitations in Mn-Mn dimer



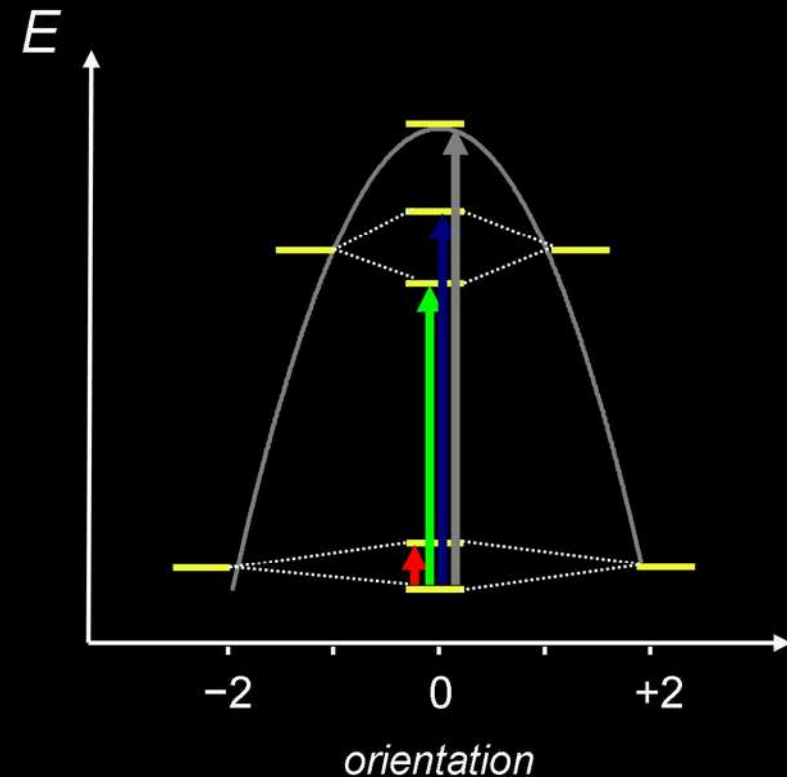
Hirjibehedin et al., Science (2006)

Fe on Cu-site on Cu₂N



Fe – Cu₂N
1 magnetic atom
 $S \approx 2$

- Same binding site as Mn
- Large uni-axial magnetic anisotropy
- Large transverse anisotropy that causes spin state mixing



$$H = -g\mu_B B_z S_z + DS_z^2 + E(S_x^2 - S_y^2)$$

$$S = 2, g = 2.11, D = -1.5 \text{ meV}, E = 0.3 \text{ meV}$$

Hirjibehedin, *et al.* Science (2007)

Exploring individual spins

Energetics:

Spin magnitude, magnetic anisotropy, Spin Hamiltonian

Science 306, 466 (2004)
Science 317, 1199 (2007)

Dynamics:

Spin lifetime, scattering mechanisms,
coherent evolution

Nature Physics, 6 340 (2010)
Science, 329 1628 (2010)

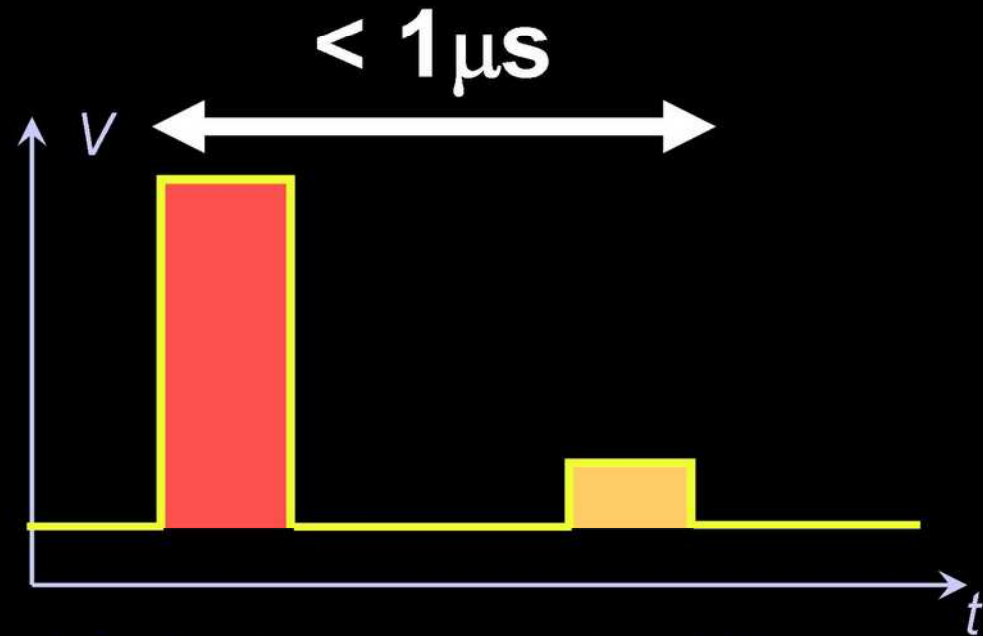
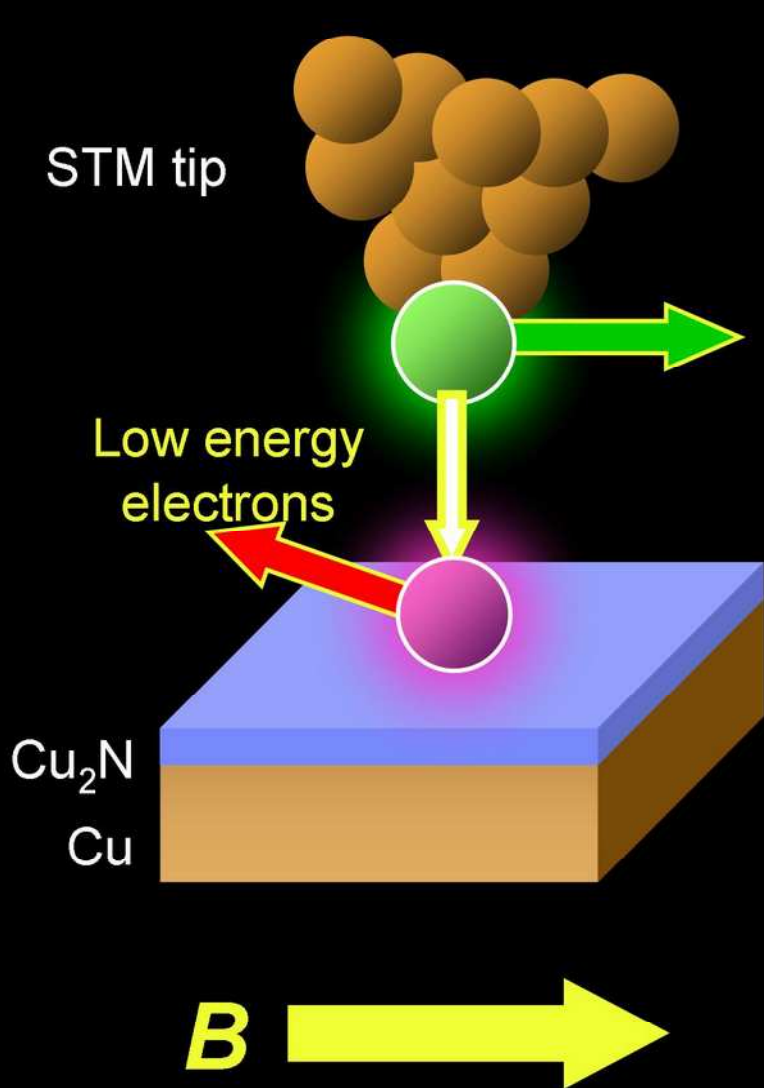


Structure:

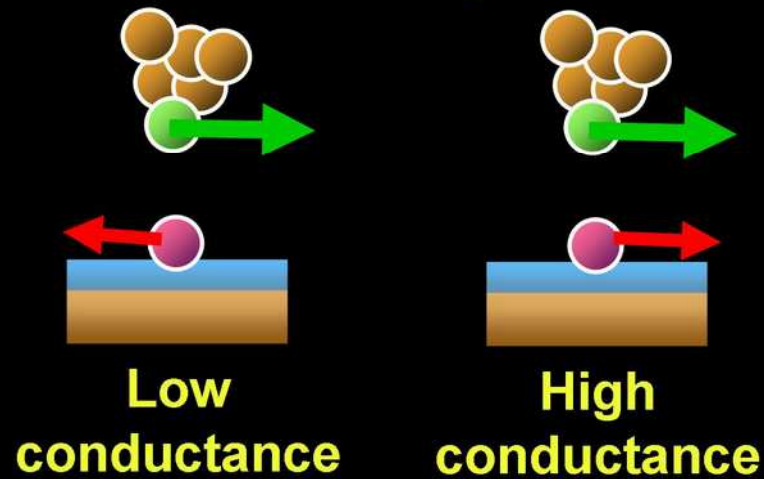
Binding site, nearby atoms, free carrier density, local environment



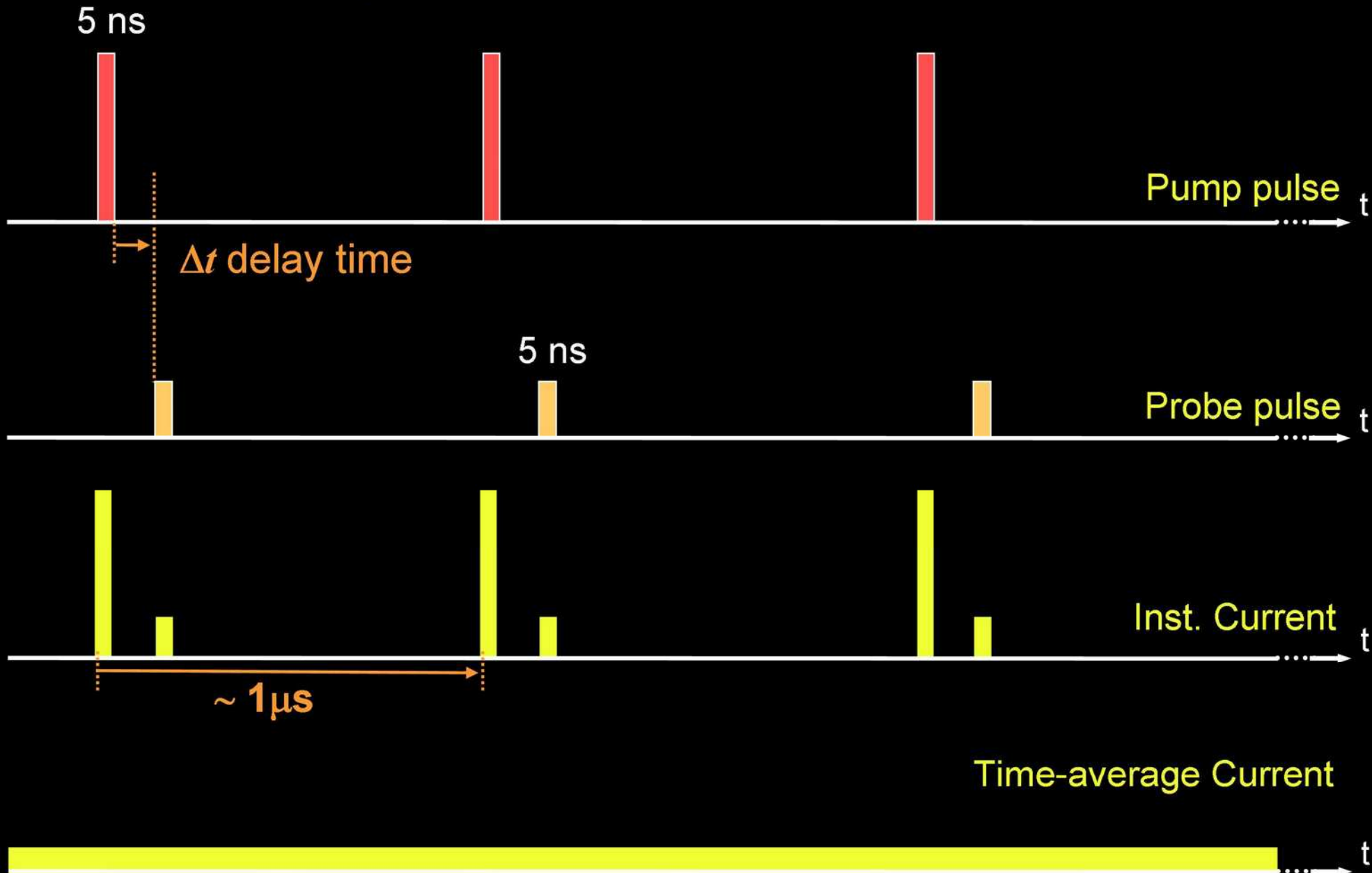
Measuring the dynamics of a single spin



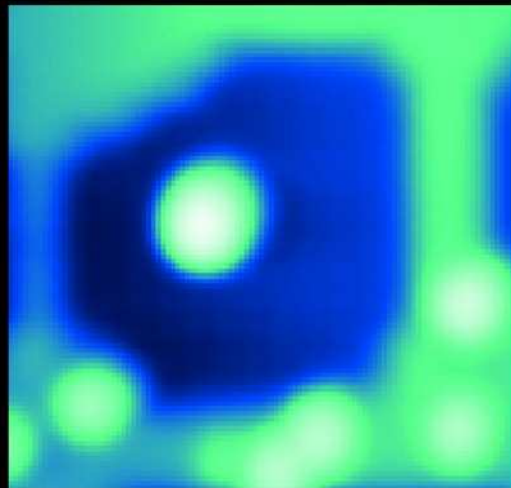
Read-out uses magnetoresistance:



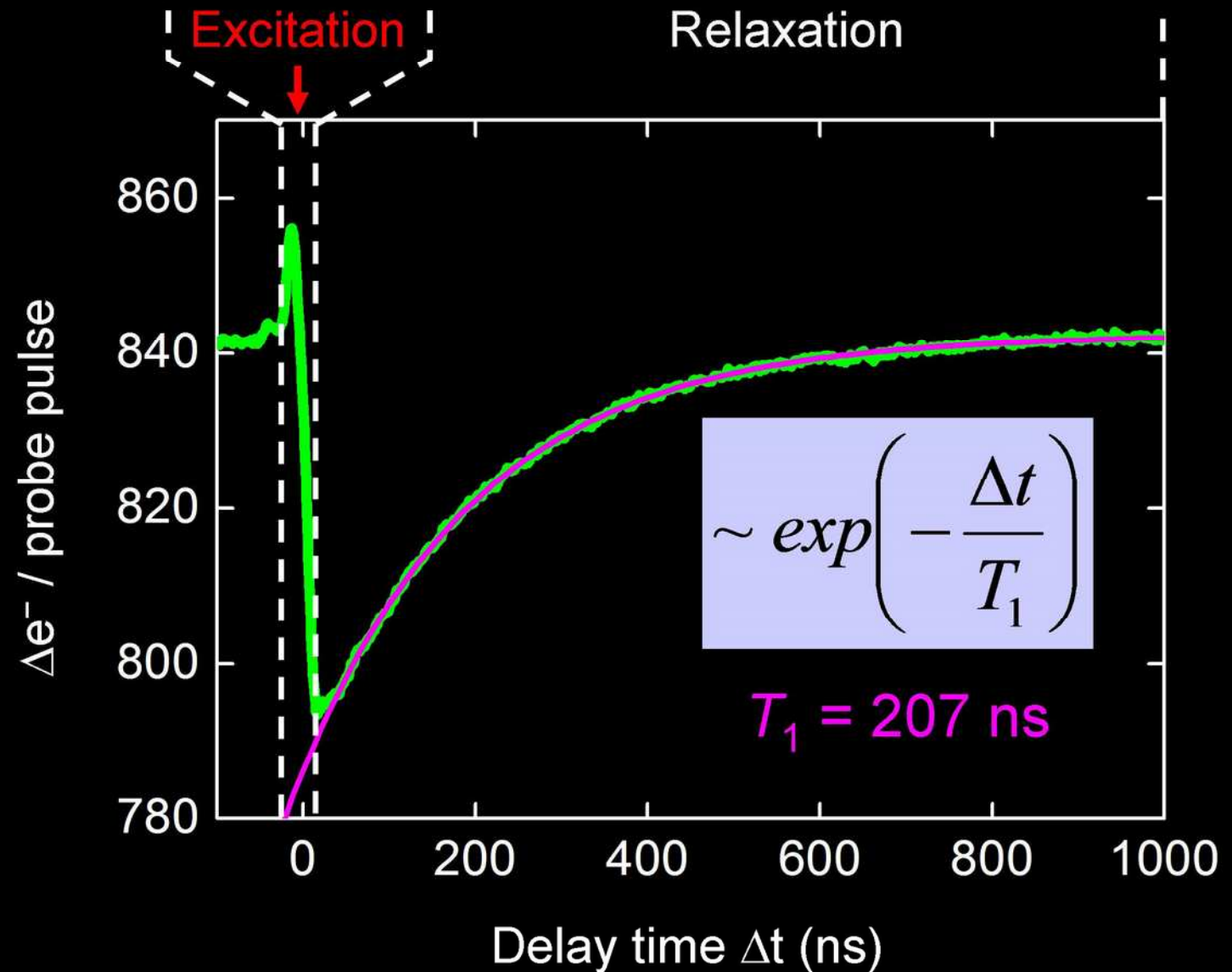
Electrical Pump – Probe scheme



Pump – Probe Measurement of a single spin

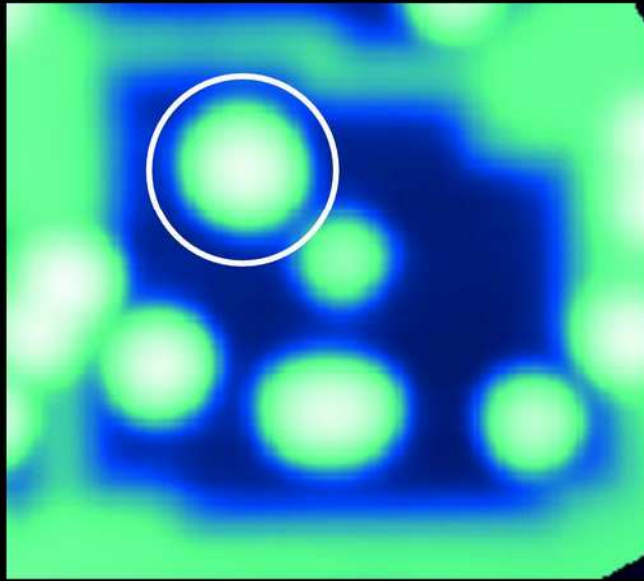


Close spaced
Fe-Cu dimer
B = 6.5T

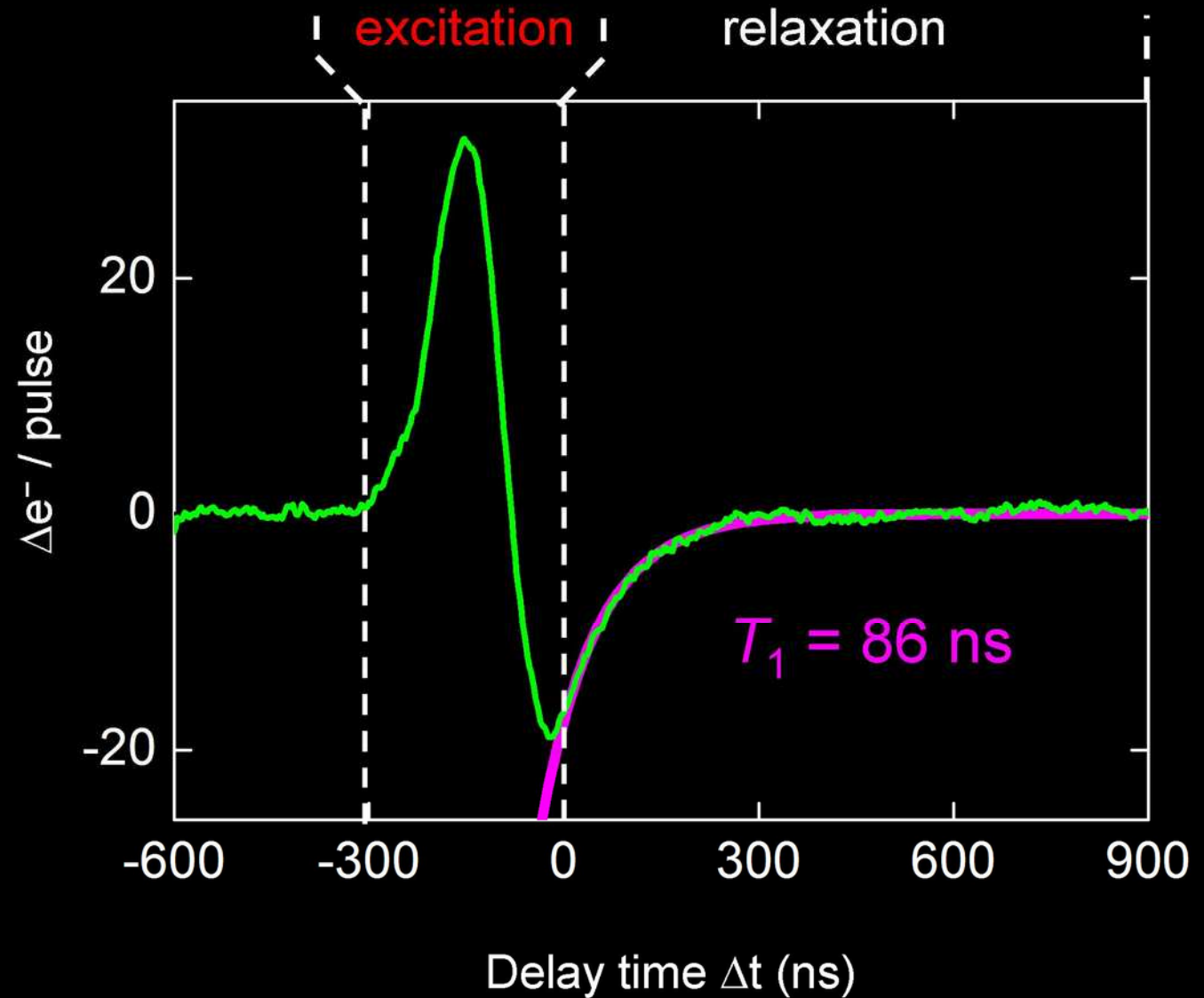


Science (2010)

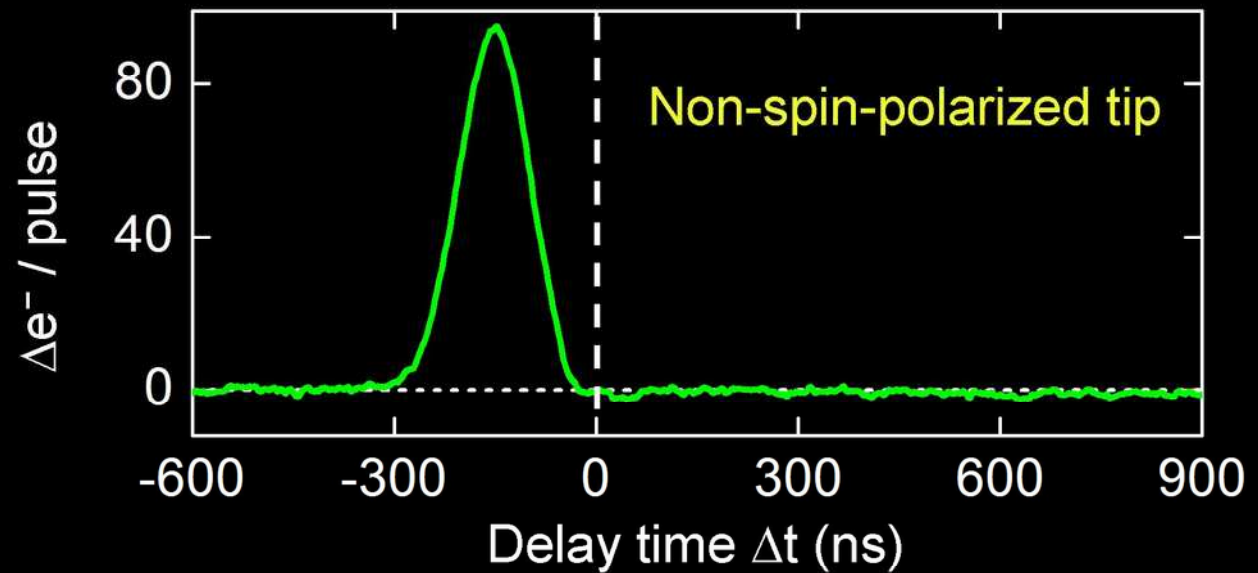
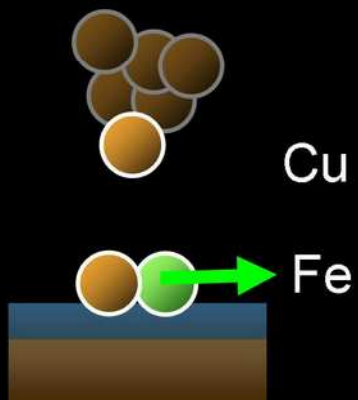
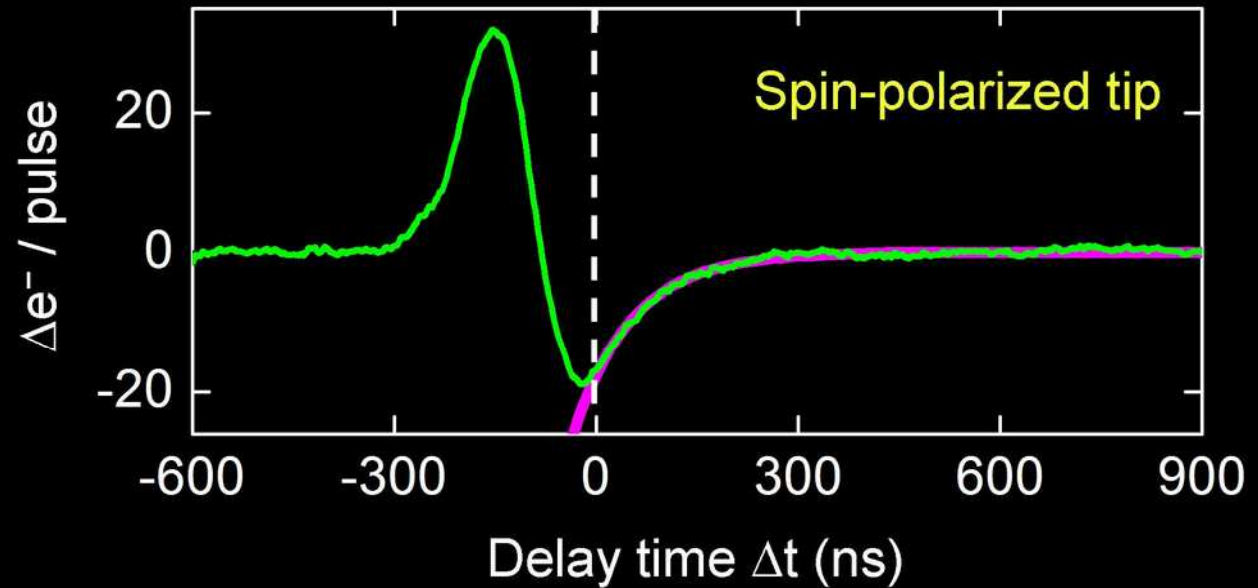
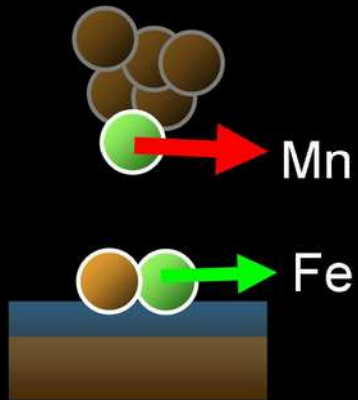
Pump – Probe Measurement of a single spin



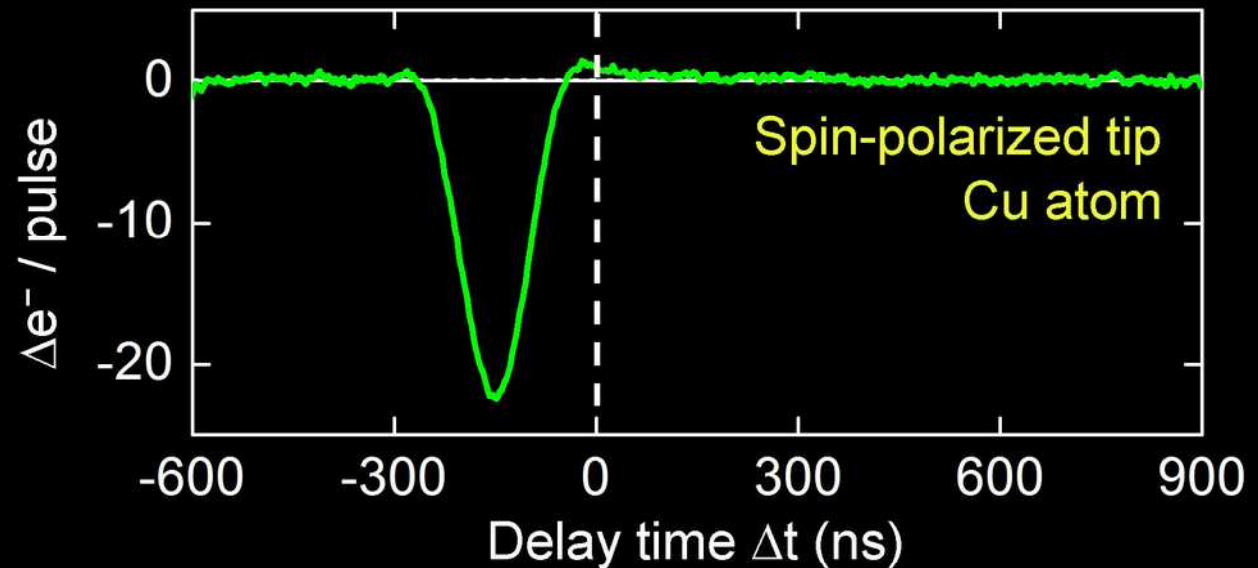
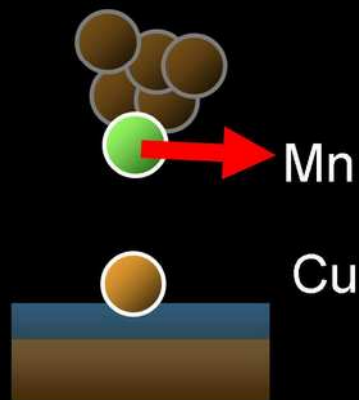
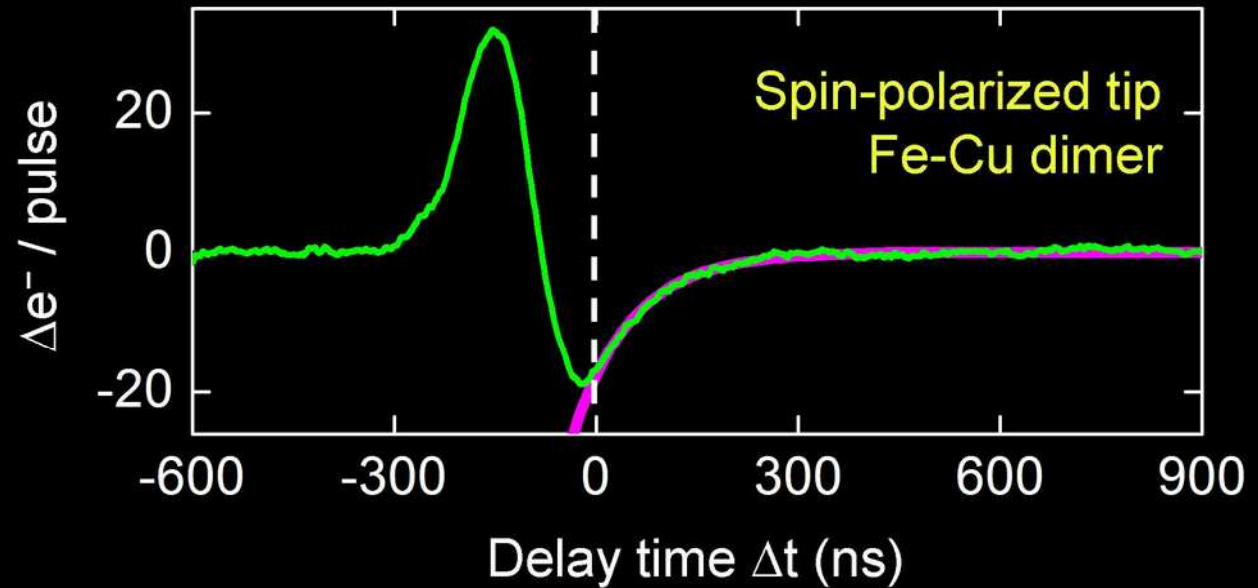
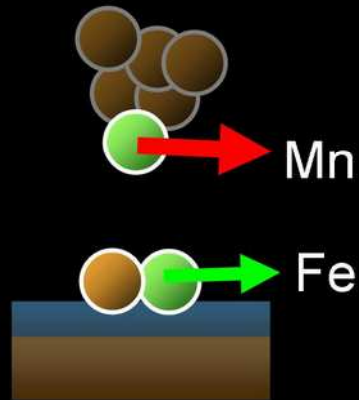
Different Fe-Cu dimer
 $B = 7\text{T}$



It is time dependent, but is it spin?

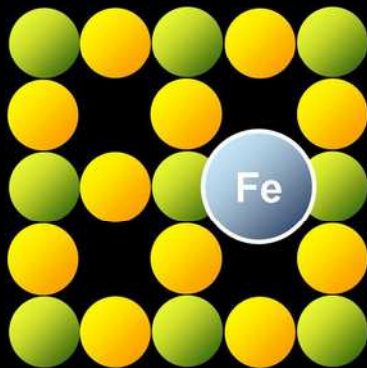
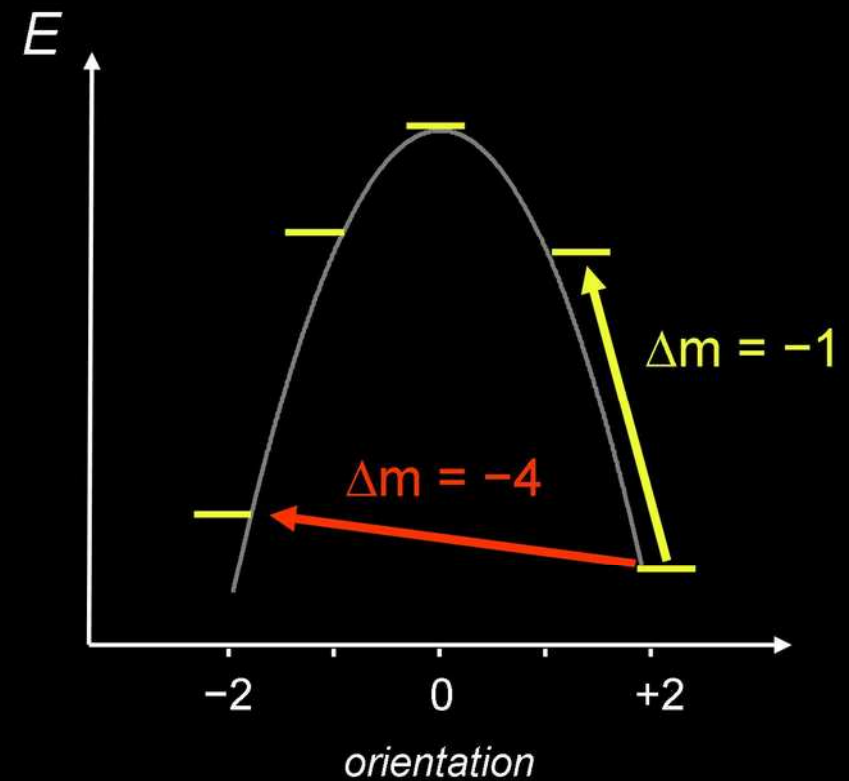
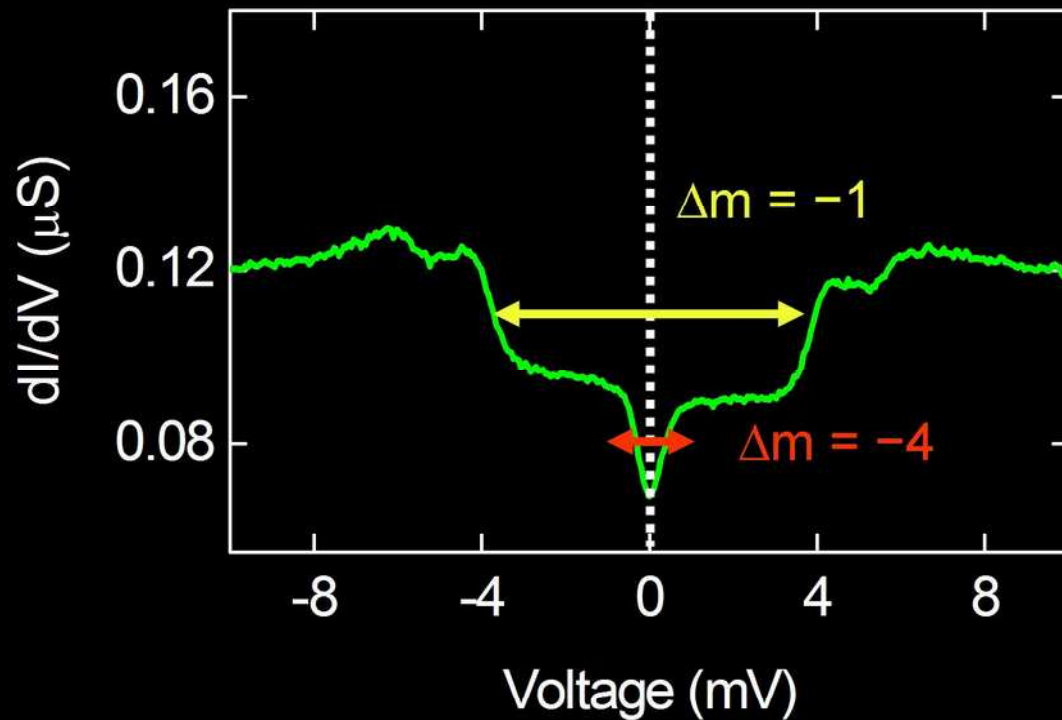


It is time dependent, but is it spin?



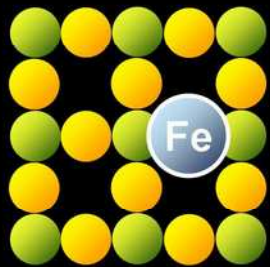
Yes, it's spin.

Anisotropy engineering

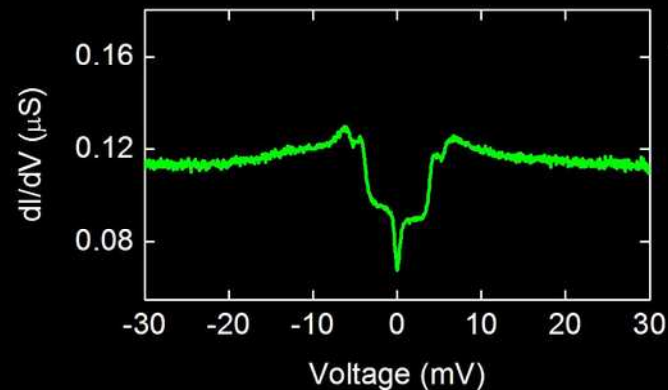


- 4 meV step: allowed transition → measure for D
- 1 meV step: forbidden transition → measure for E
- Goal: maximize D and minimize E.

Anisotropy engineering

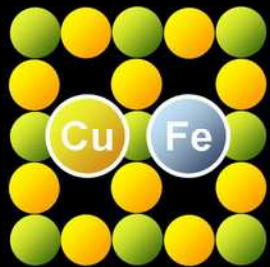


Fe

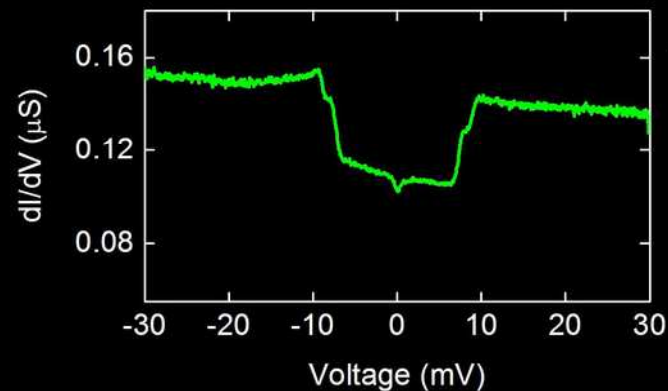


Anisotropy barrier: 5 meV
Strong mixing

Unstable. $T_1 \approx 1$ ns

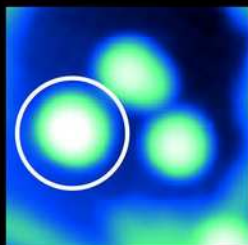


Cu-N-Fe

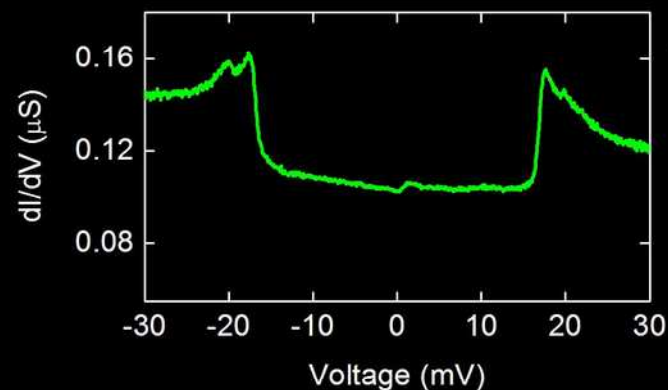


Anisotropy barrier: 10 meV
Reduced mixing

Increased stability. $T_1 \approx 8$ ns



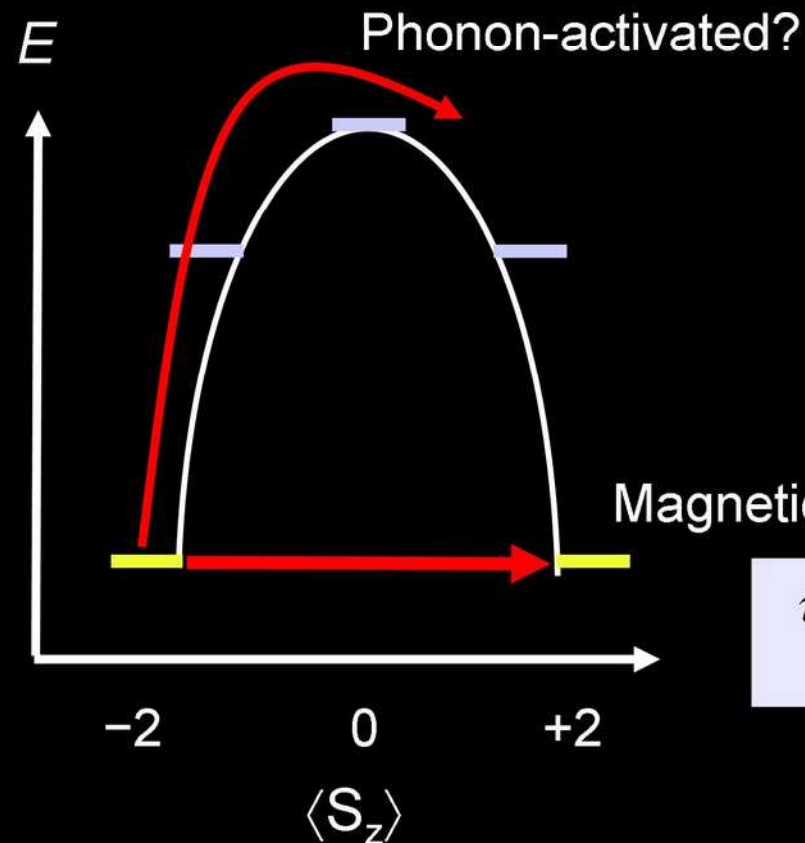
Close-spaced
Cu-Fe dimer



Anisotropy barrier: 22 meV
Minimal mixing

Long lived. $T_1 > 200$ ns

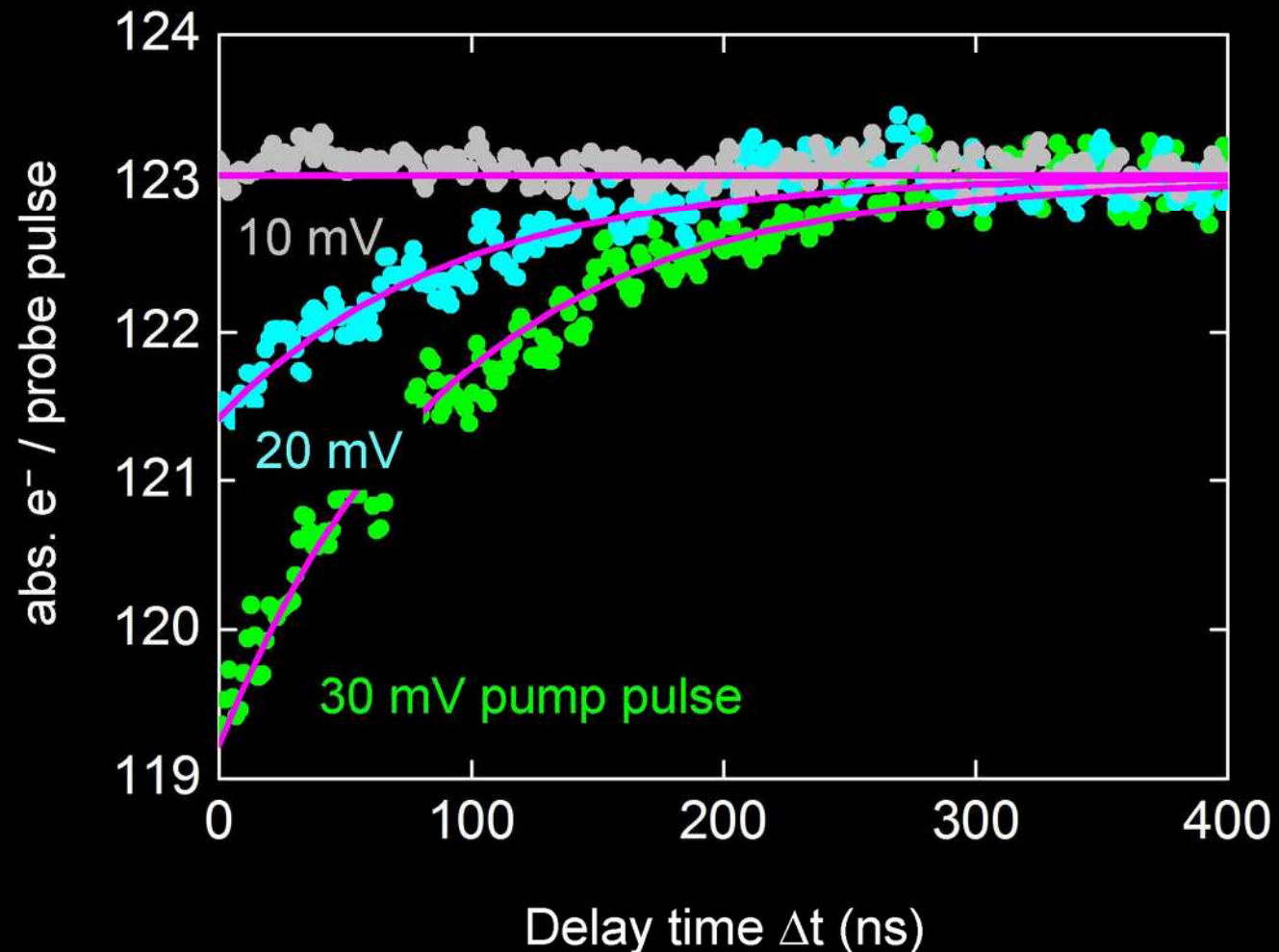
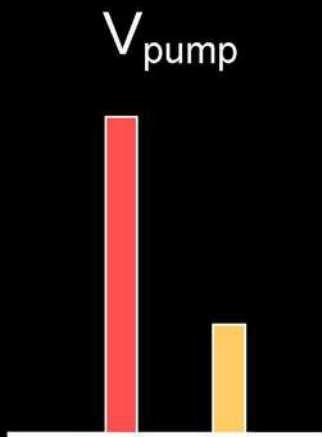
What is the spin relaxation channel?



$$\tau_1 \sim \exp\left(\frac{E_a}{k_B T}\right)$$

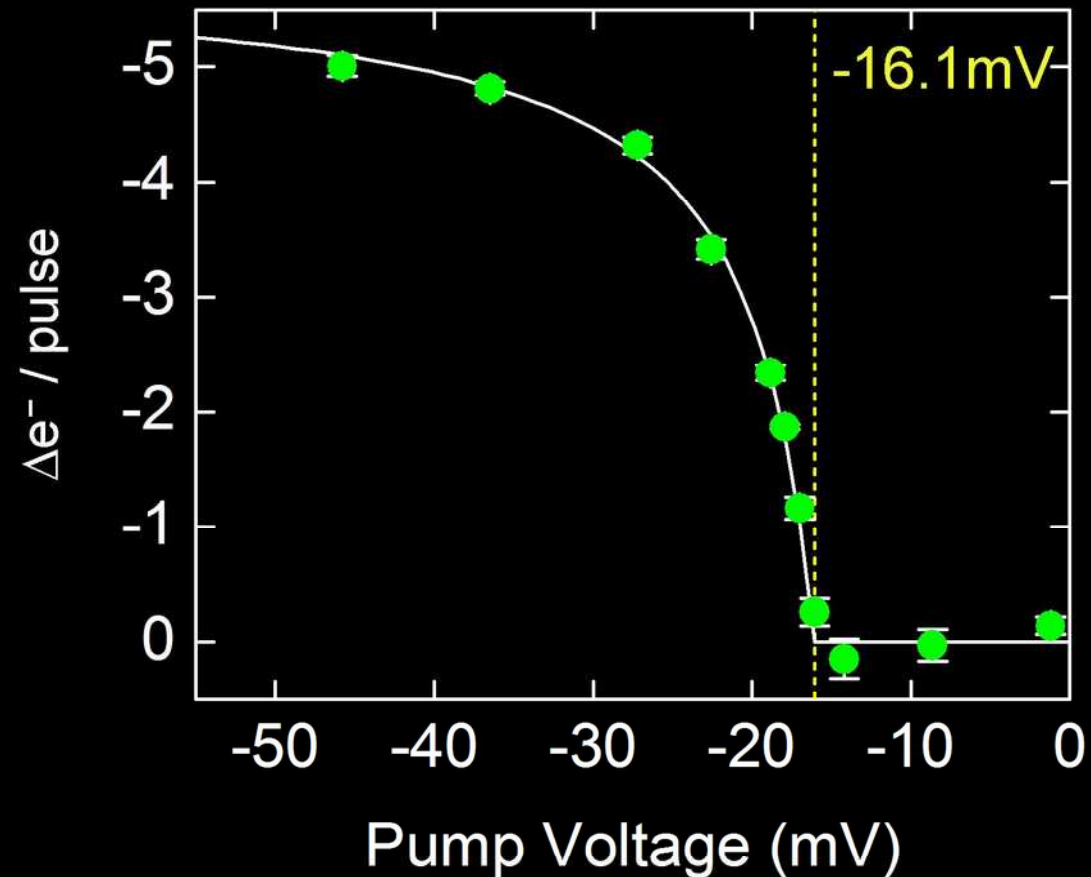
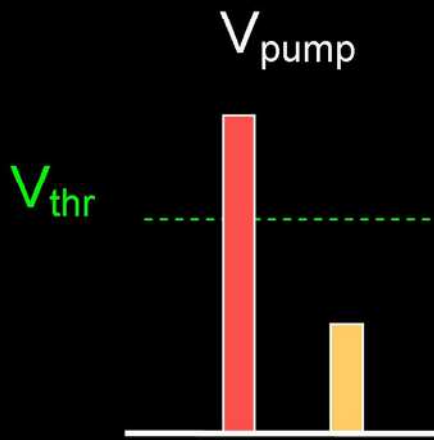
τ_1 strongly dependent
on magnetic field

Energy dependence of the excitation



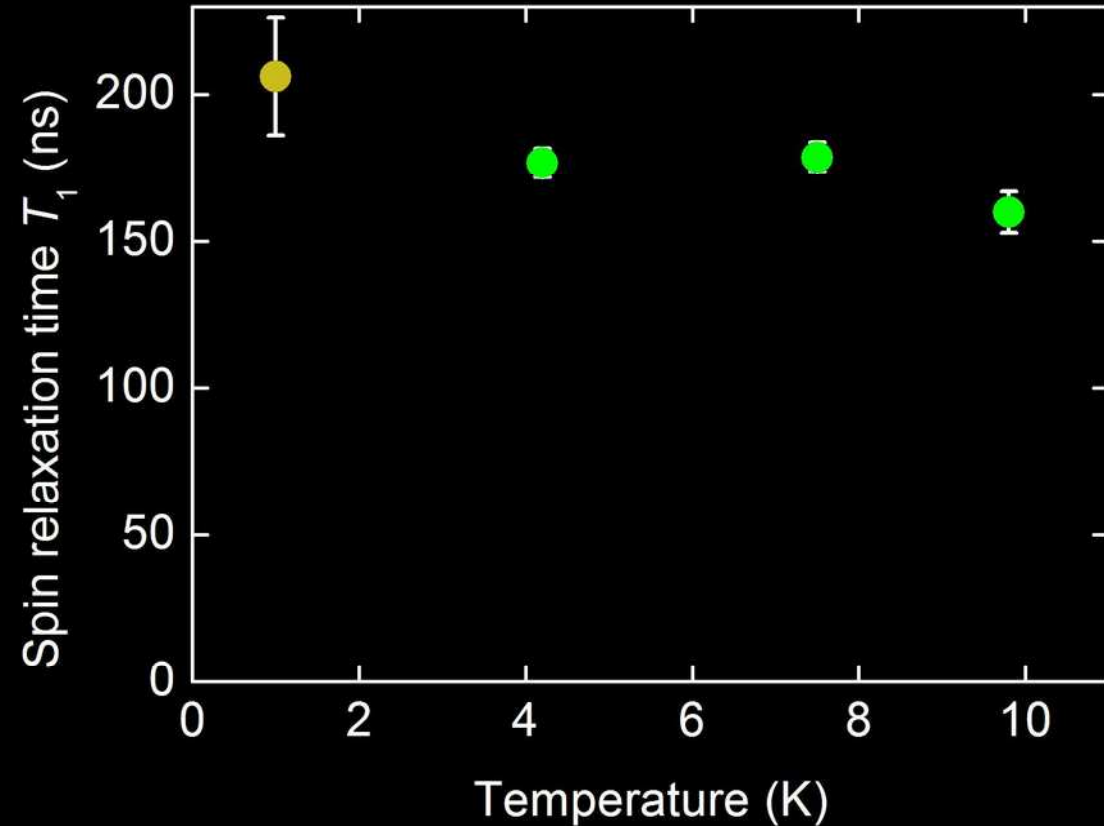
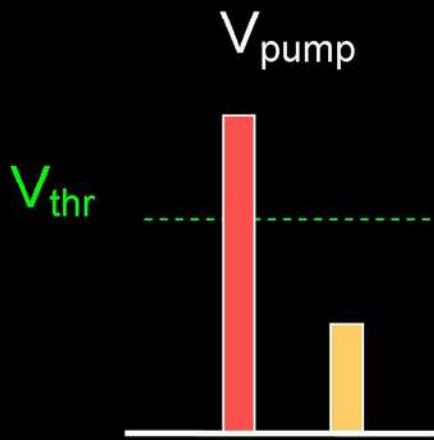
- **Signal magnitude changes with the energy of the pump pulse**
- **Characteristic relaxation time stays constant.**

Energy dependence of pump-probe signal



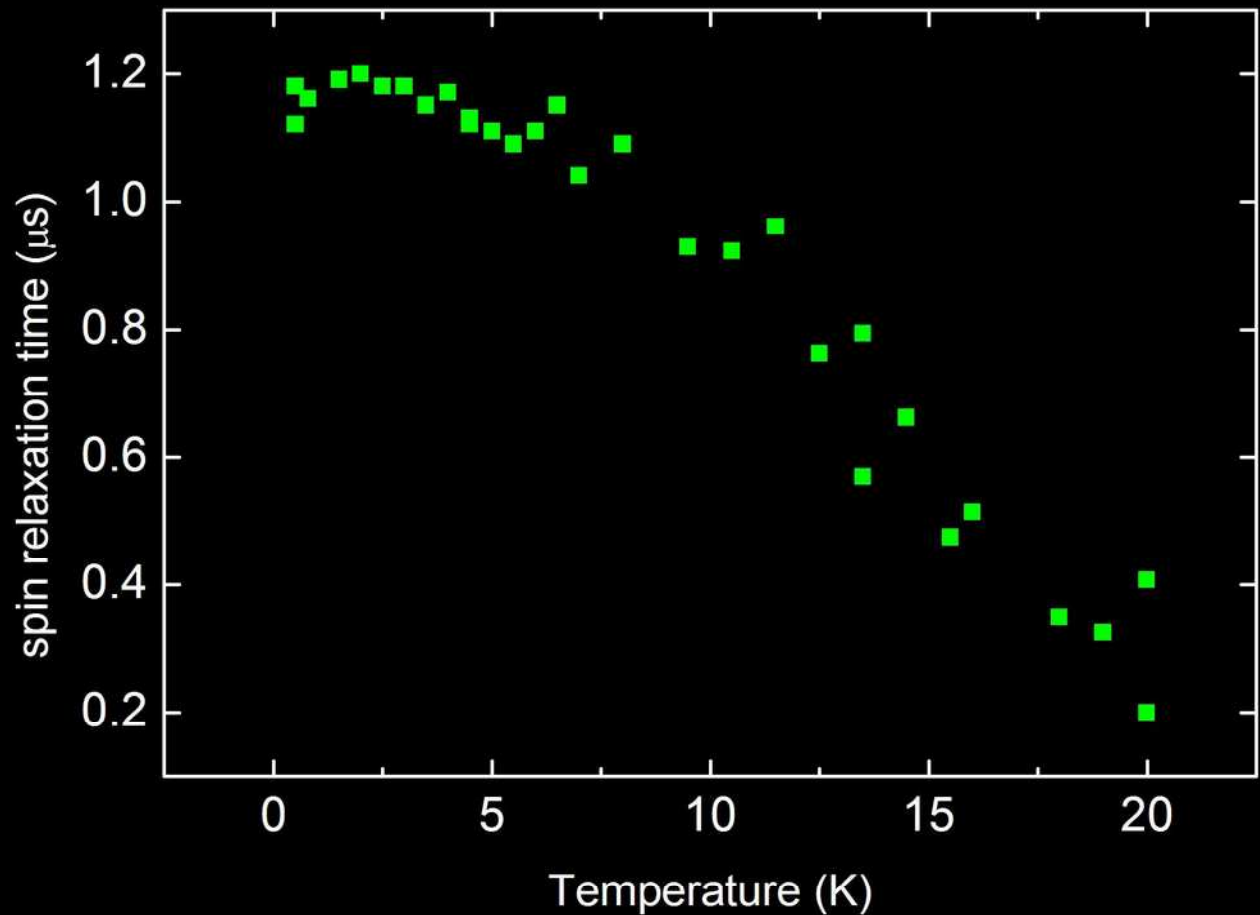
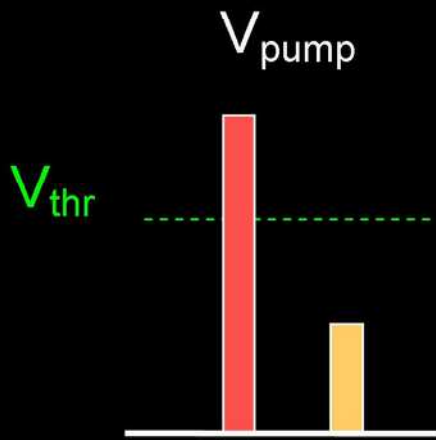
- **One clearly defined threshold energy of 16 meV**
 - ❖ **Thermal energy at 0.5 K is 40 μeV**

Energy dependence of pump-probe signal



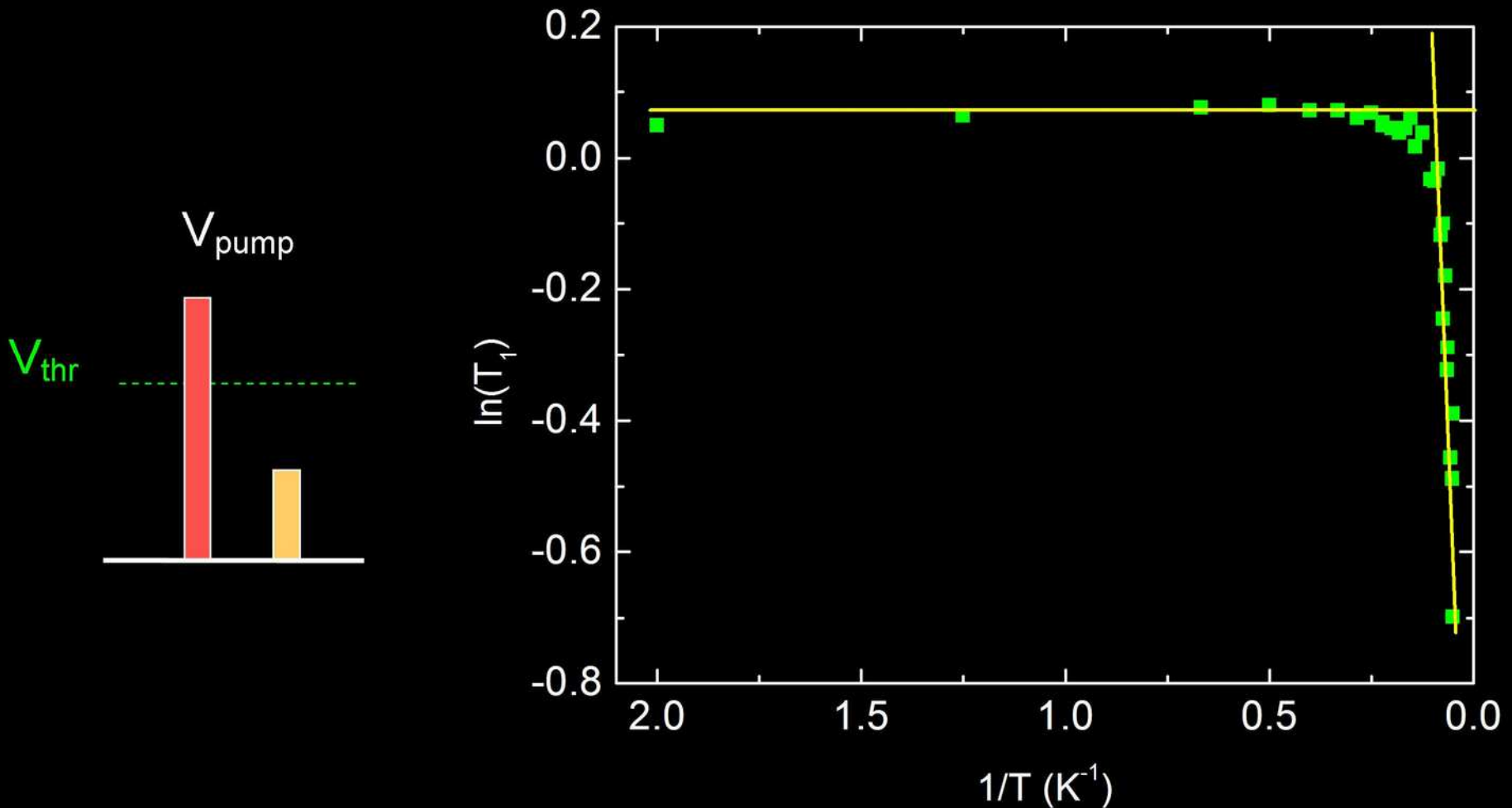
- **One clearly defined threshold energy of 16 meV**
 - ❖ **Thermal energy at 0.5 K is 40 μ eV**

Energy dependence of pump-probe signal



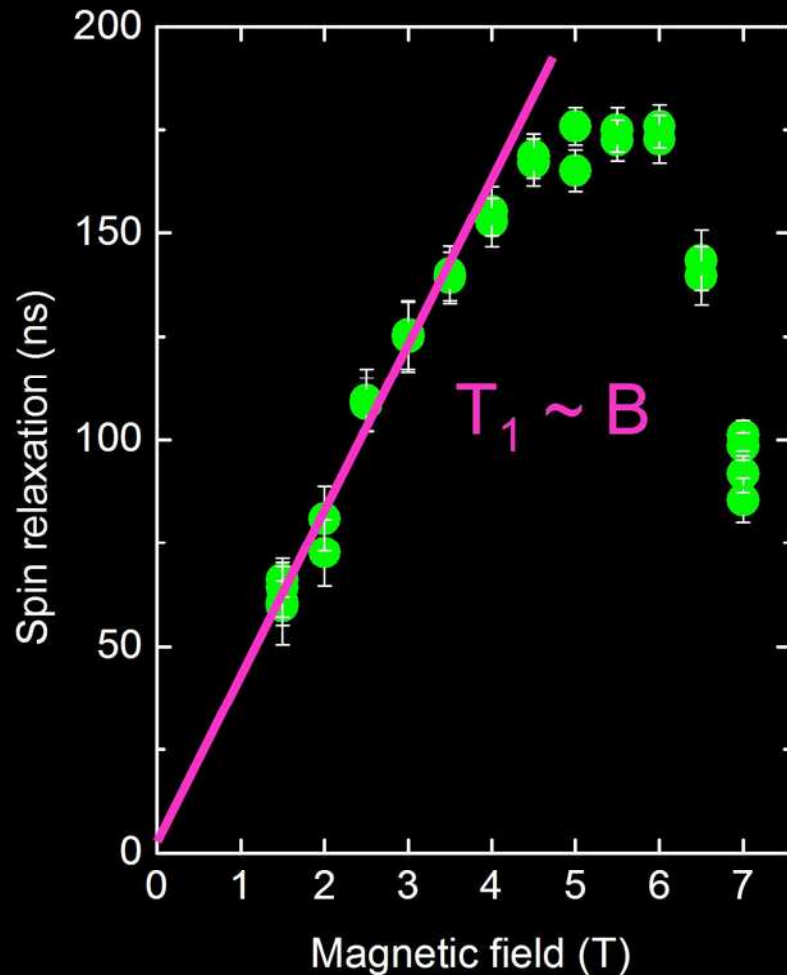
- **One clearly defined threshold energy of 16 meV**
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Energy dependence of pump-probe signal

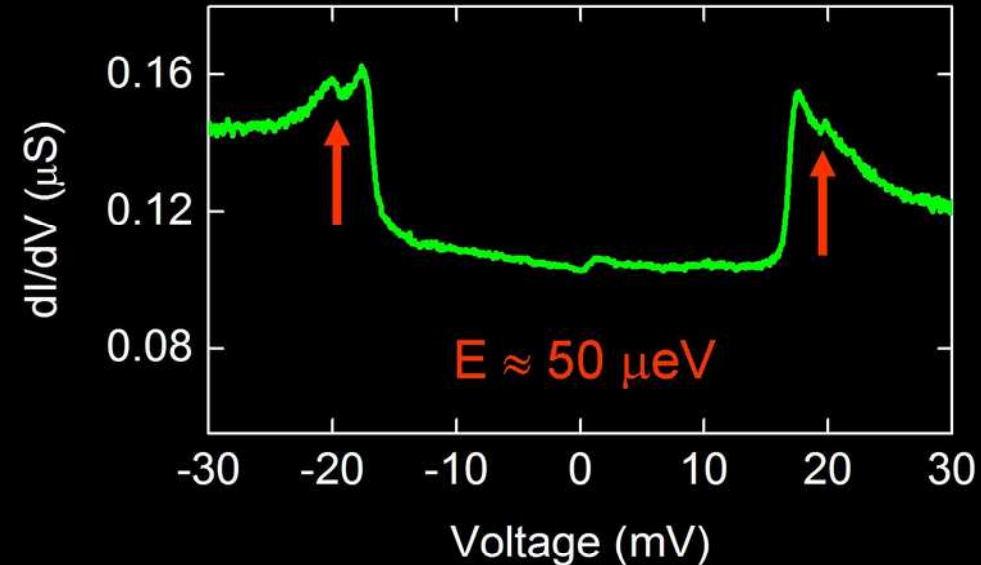


- **One clearly defined threshold energy of 16 meV**
 - ❖ Thermal energy at 0.5 K is 40 μeV
- **Thermal relaxation negligible**

Magnetic field dependence of spin relaxation time

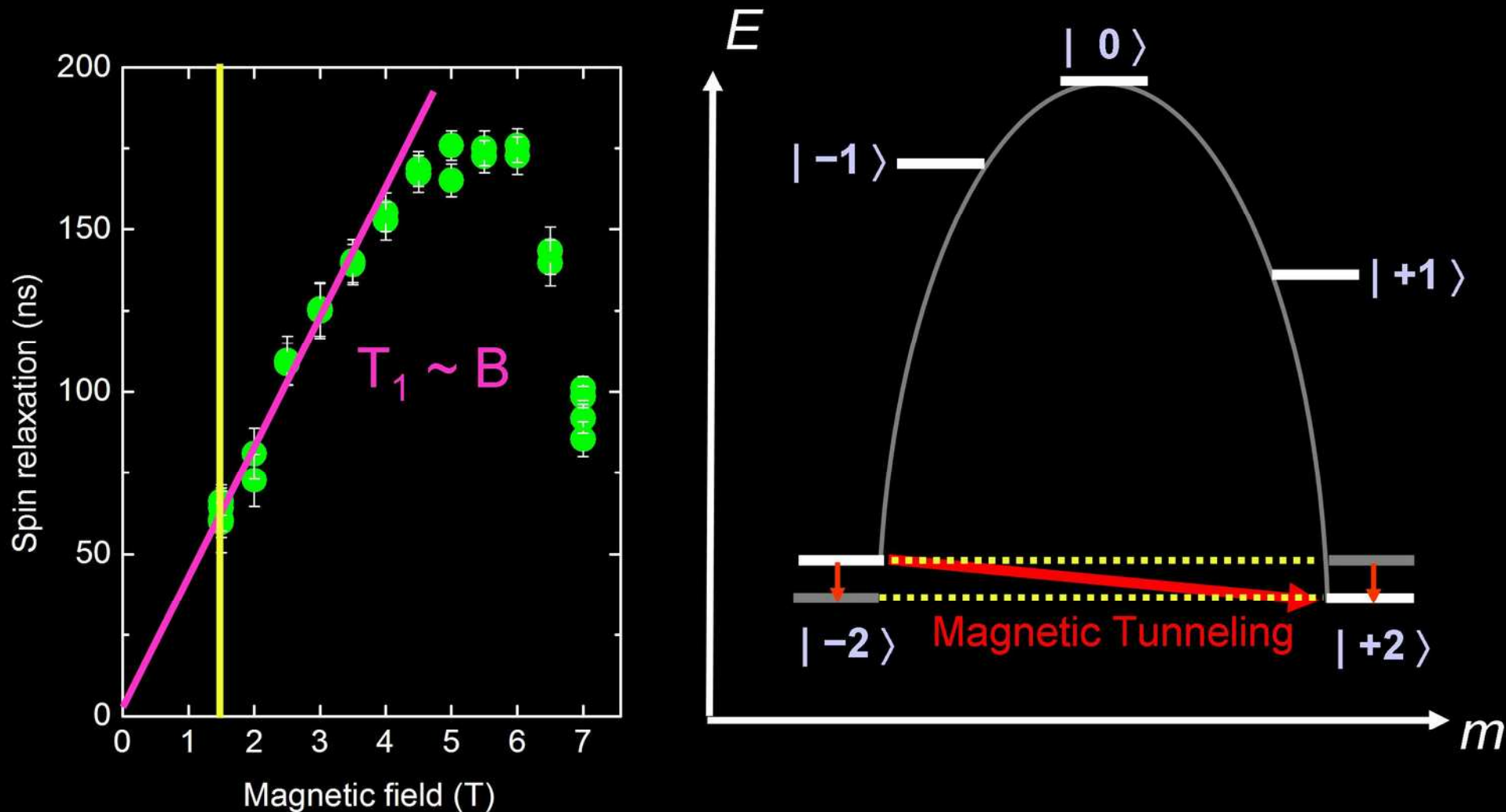


- **Dynamical information:** consistent with magnetic tunneling limiting the spin lifetime.



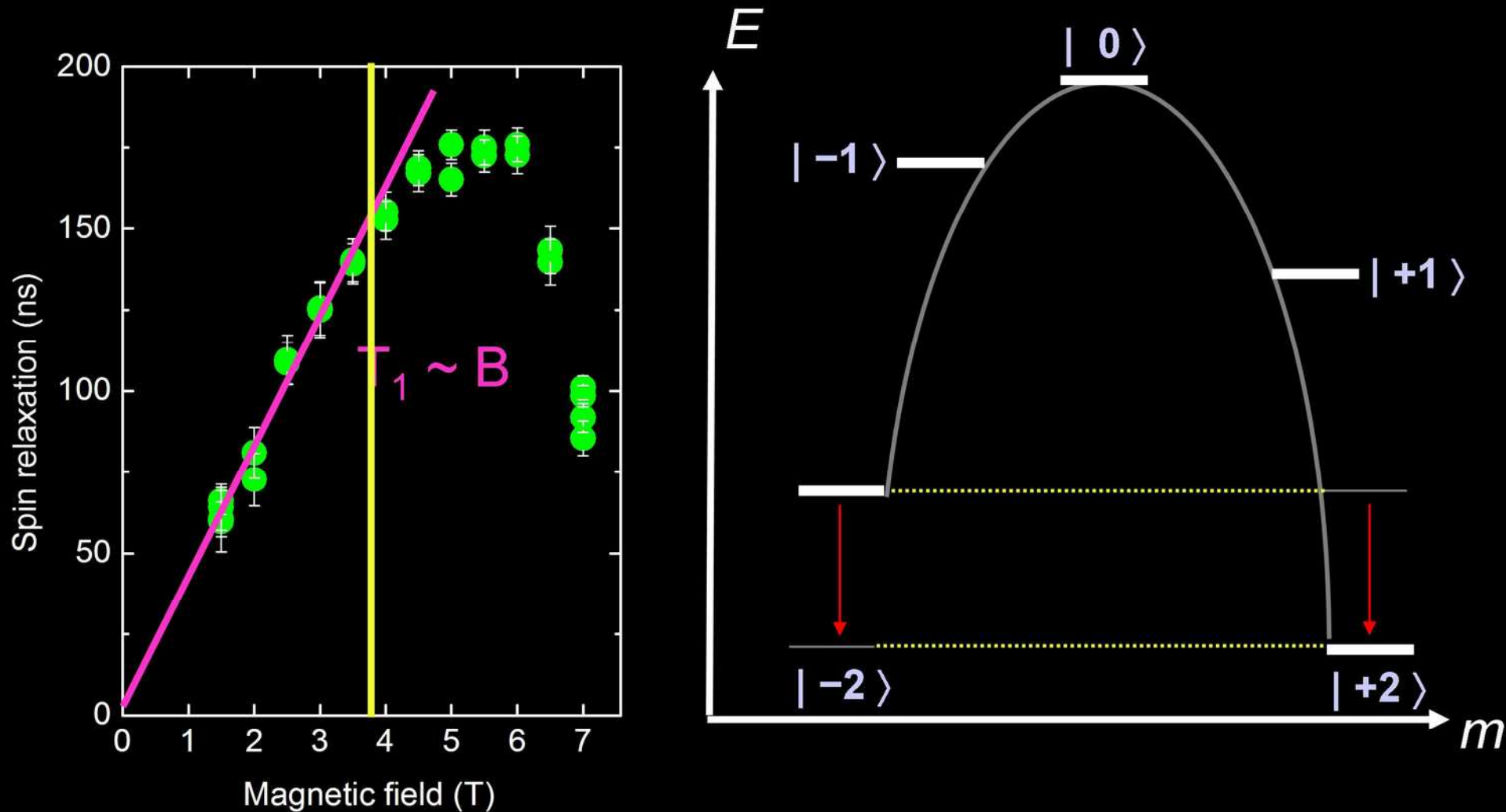
- **Energetic information:** magnetic tunneling caused by finite transverse anisotropy

Magnetic field dependence of spin relaxation time



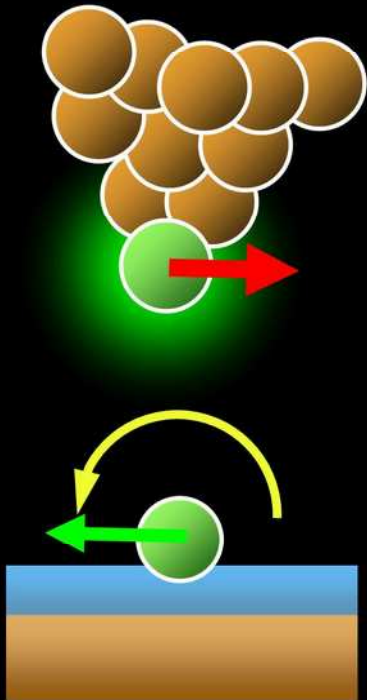
➤ **Relaxation arises from mixing of the $| -2 \rangle$ and the $| +2 \rangle$ states**

Magnetic field dependence of spin relaxation time



- **Relaxation arises from mixing of the $| -2 \rangle$ and the $| +2 \rangle$ states**
- **The mixing decreases with increasing magnetic field**

Summary



STM-based exploration of quantum spins:

- Energetic structure
- Dynamic behavior
- Spin lifetime engineering

(ns × nm)