Spin decoherence at high magnetic fields

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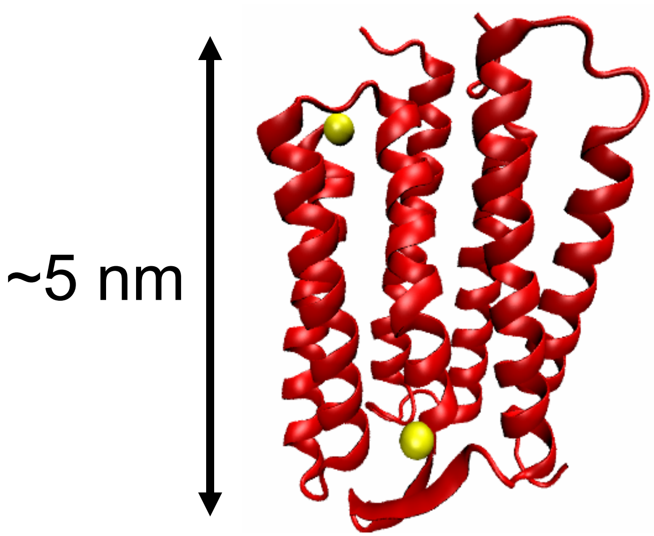
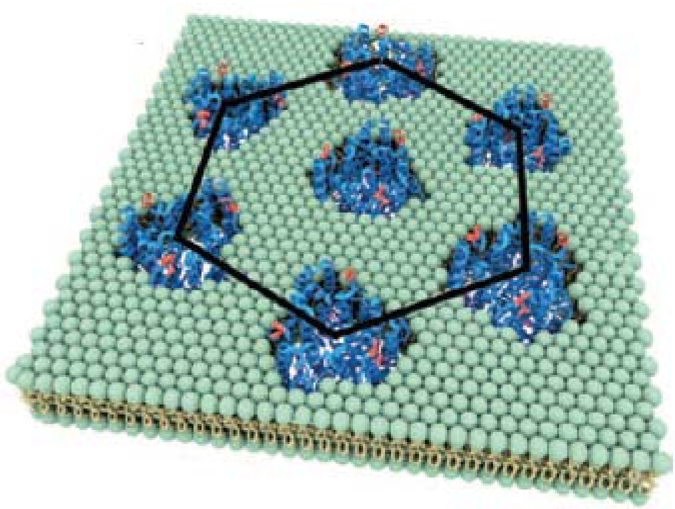
USC

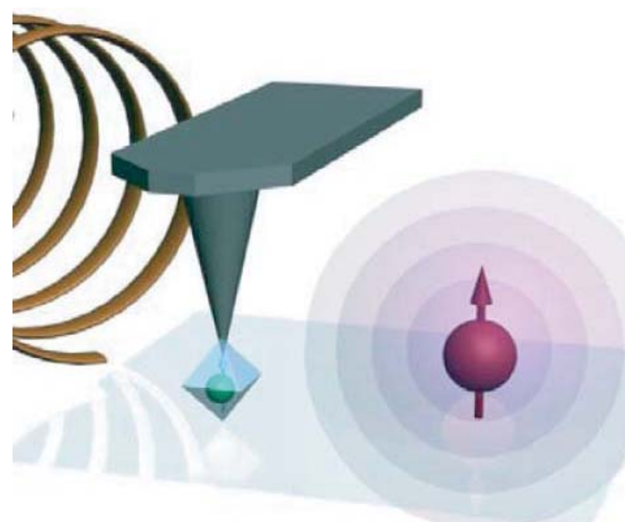
Distance measurement using EPR



- Large biomacromolecules but cannot be crystallized easily, *e.g.* membrane proteins.
- Assembly of molecules
- Electron spin based magnetic sensor

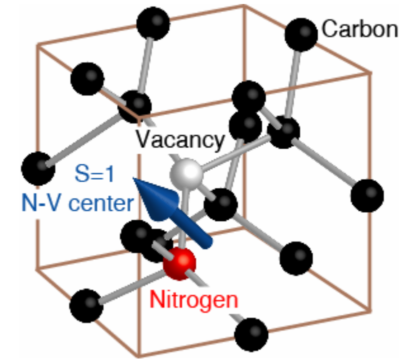


H. Liang *et al.*, *PNAS* **104**, 8212 (07)

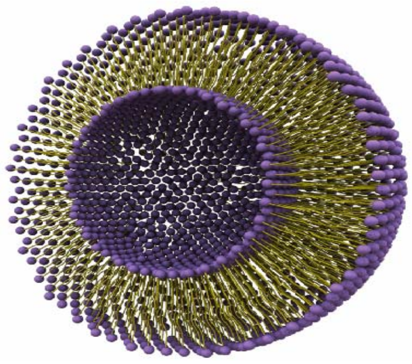


G. Balasubramanian *et al.*, *Nature* **455**, 648 (08)

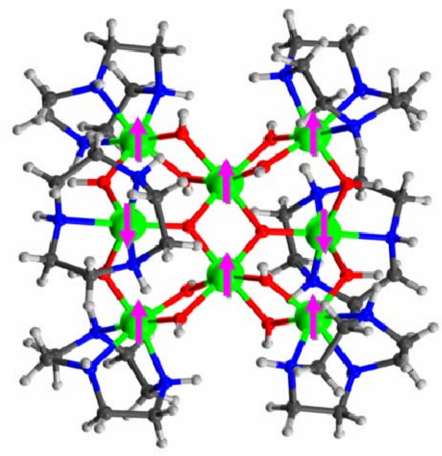
Outline



1. Quenching spin decoherence of NV centers in diamond



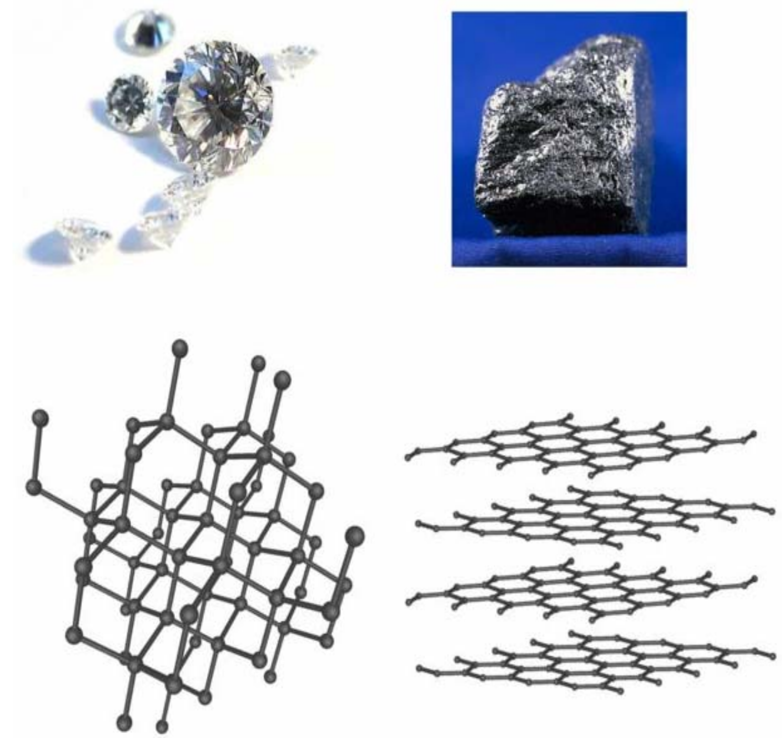
2. T_2 based distance measurement



3. Spin decoherence of Fe8 single-molecule magnets

Diamond and impurities

- Hardest material
- Excellent thermal conductor
- A diamond is a crystal of tetrahedrally bonded carbon atoms.
- Diamond is classified by impurity contents.

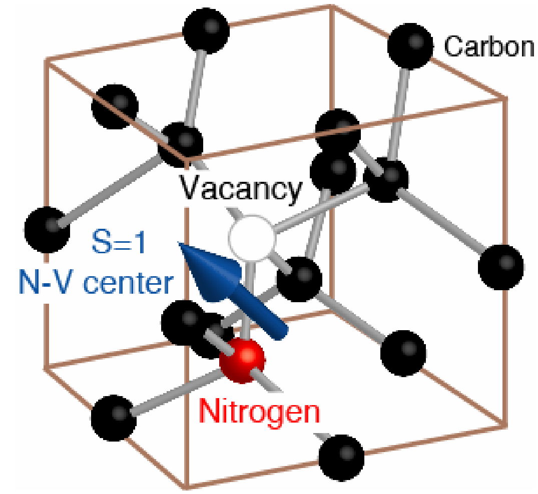


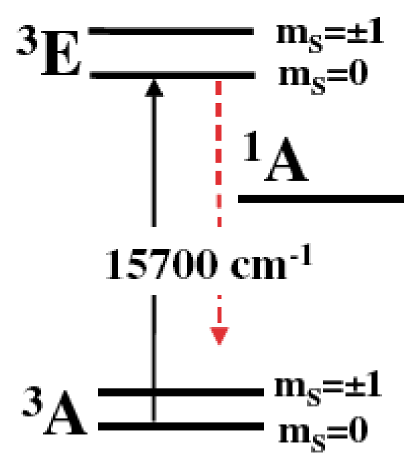
Diamond Graphite

Type	Ia	Ib	IIa	IIb
Natural abundance	~98 %	~0.1 %	1~2 %	~0 %
Nitrogen (ppm)	~2 x 10^3	1~10^2	~1	~1
Others (ppm)				~100 (B)
Color	Clear~Yellow	Yellow	Clear	Blue

NV centers in Diamond

- Rapid spin polarization
- Single spin read-out
- Long spin coherence time at room temperature

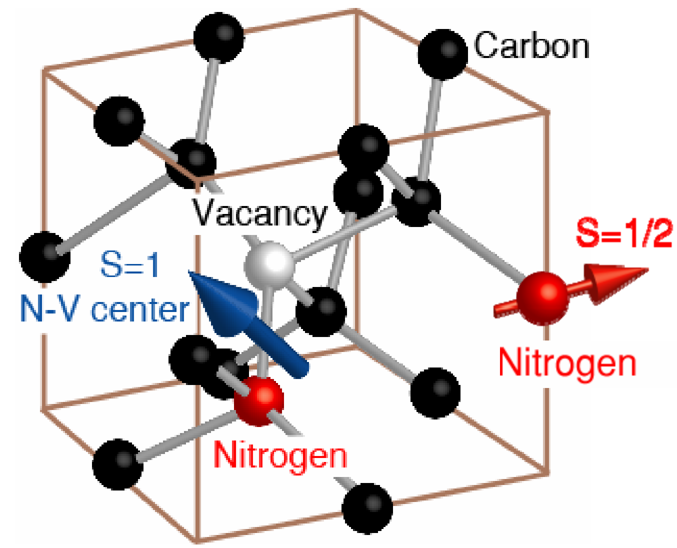


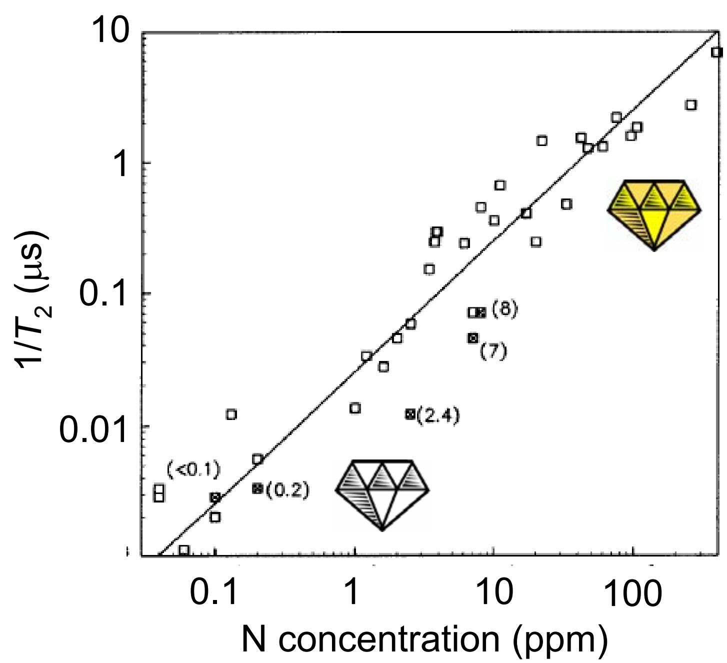


$$H_{NV} = g\mu_B \mathbf{B}\cdot\mathbf{S} + D(S_z^2 + 1/3S(S+1))$$

Decoherence of NV center

- N electron spin flip-flops
  - J. A. van Wyk *et al.*, *J. Phys. D: Appl. Phys.* **30**, 1790 (1997).



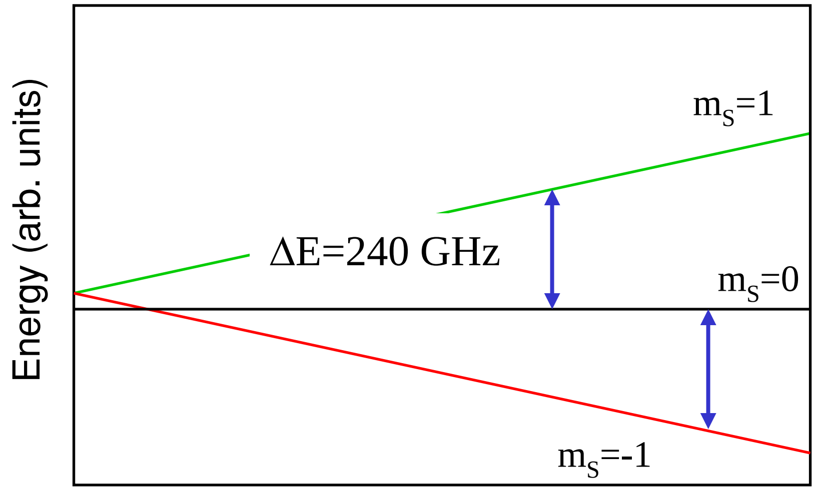


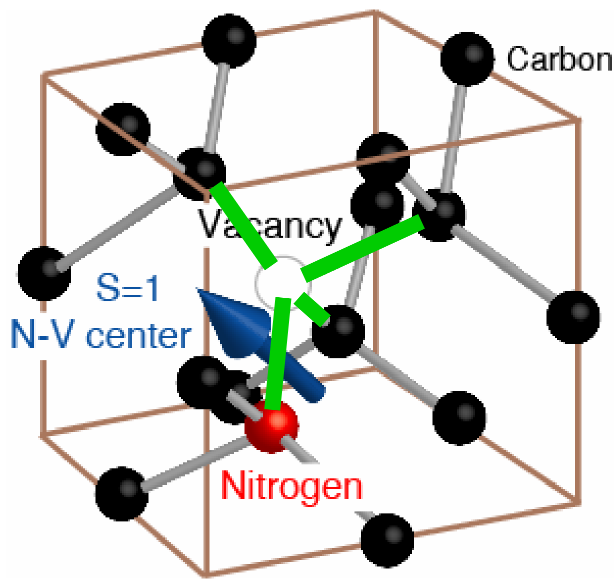
Continuous wave EPR

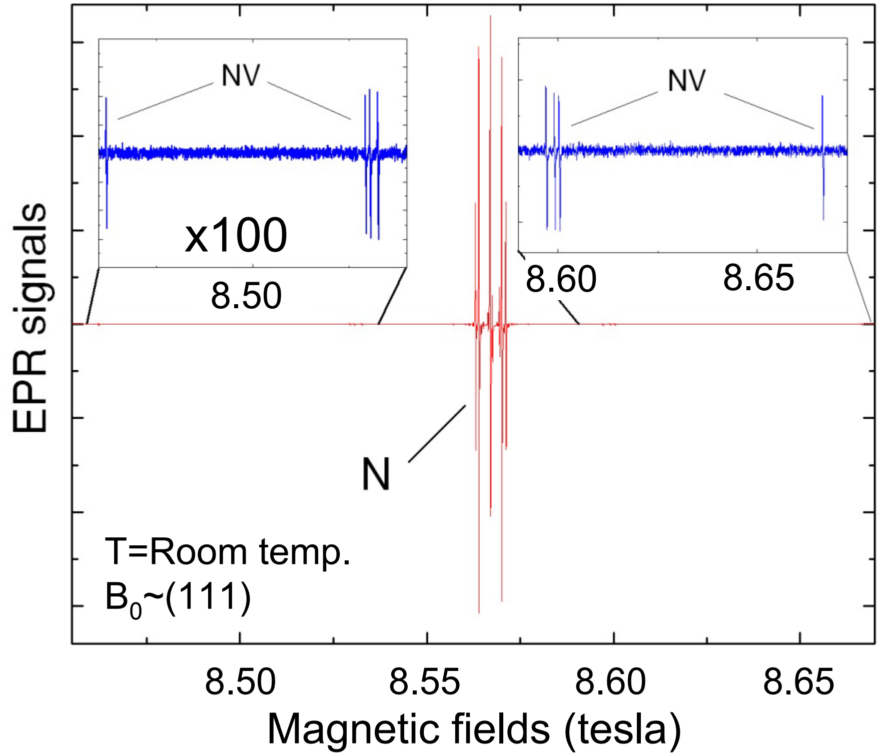
- NV center (S=1)

$$H_{NV} = g\mu_B \mathbf{B}\cdot\mathbf{S} + D(S_z^2 + 1/3S(S+1))$$

  – Zero-field splitting

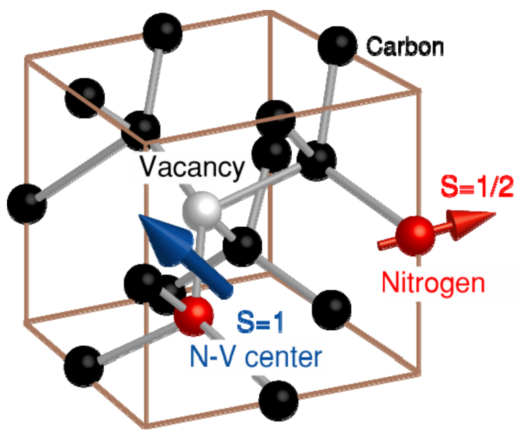


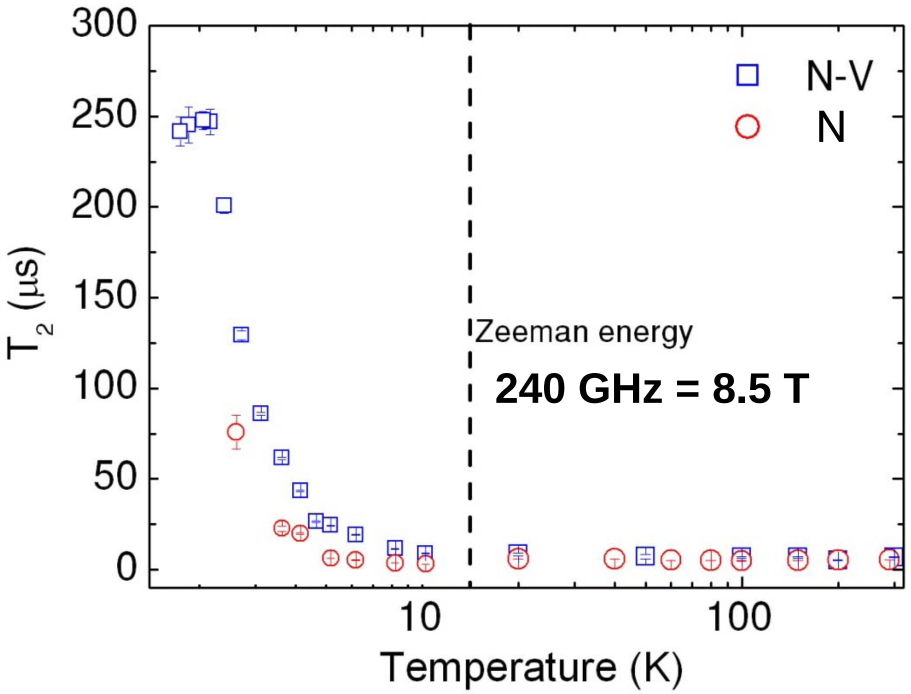




T_2 of NV centers in diamond

- Temperature dependence of T_2 for NV centers in diamond



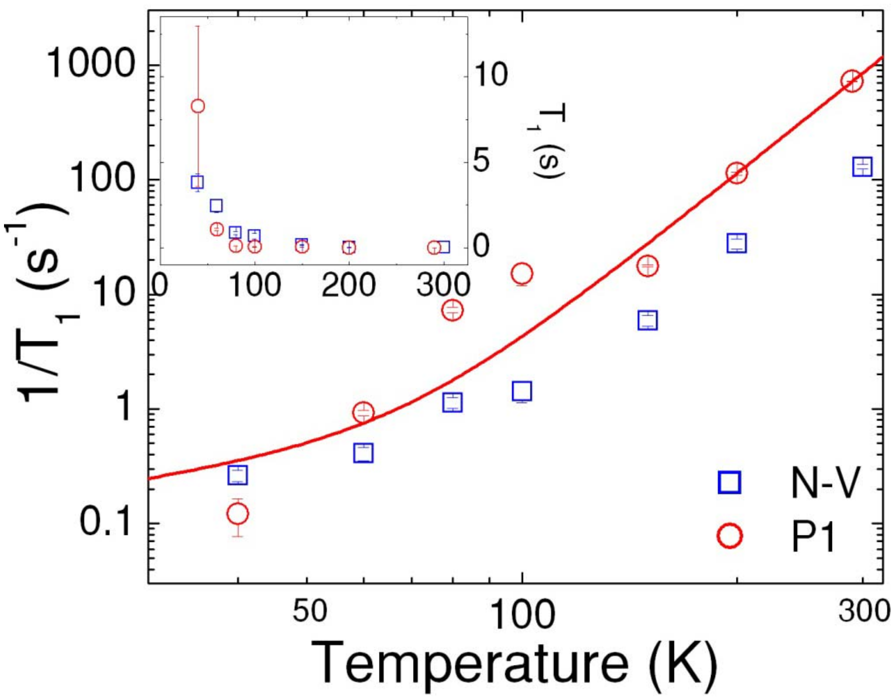


- N-V:
  - T>11.5 K : T_2 = 6.7 μs → 8.3 μs
  - T < 2 K : T_2 ~ 250 μs

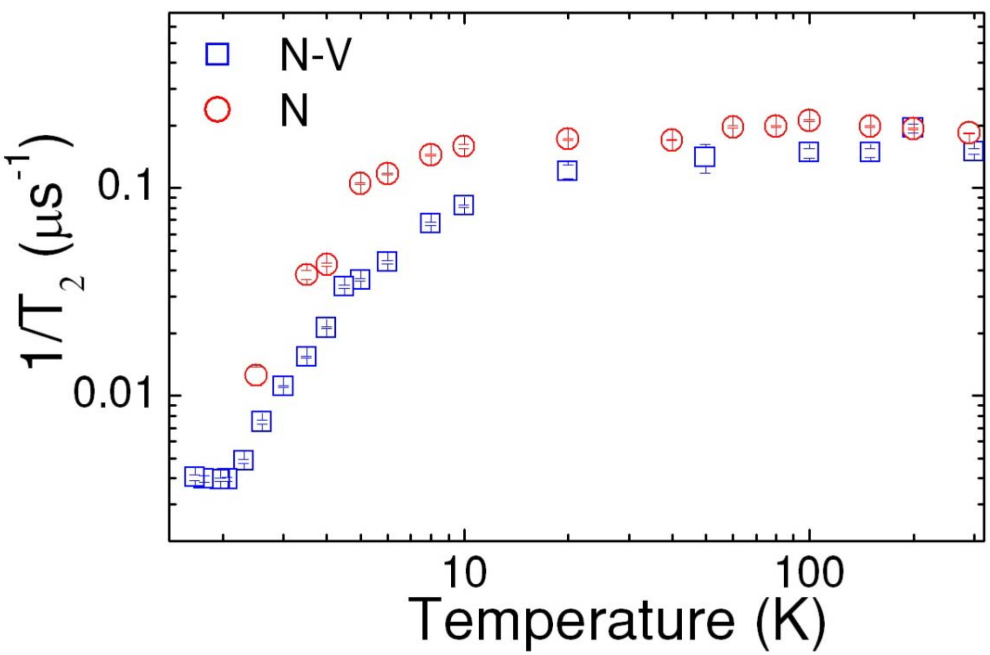
- N:
  - T>11.5 K : T_2 = 5.5 μs → 5.8 μs
  - T = 2.5 K : T_2 ~ 80 μs

Quenching spin bath decoherence

T_1



T_2

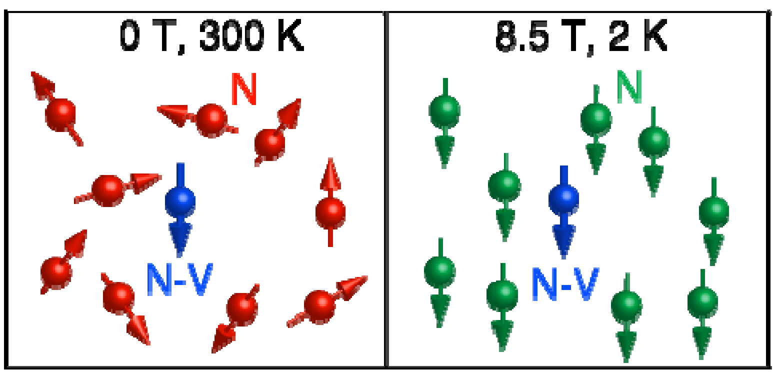


Quenching spin bath decoherence

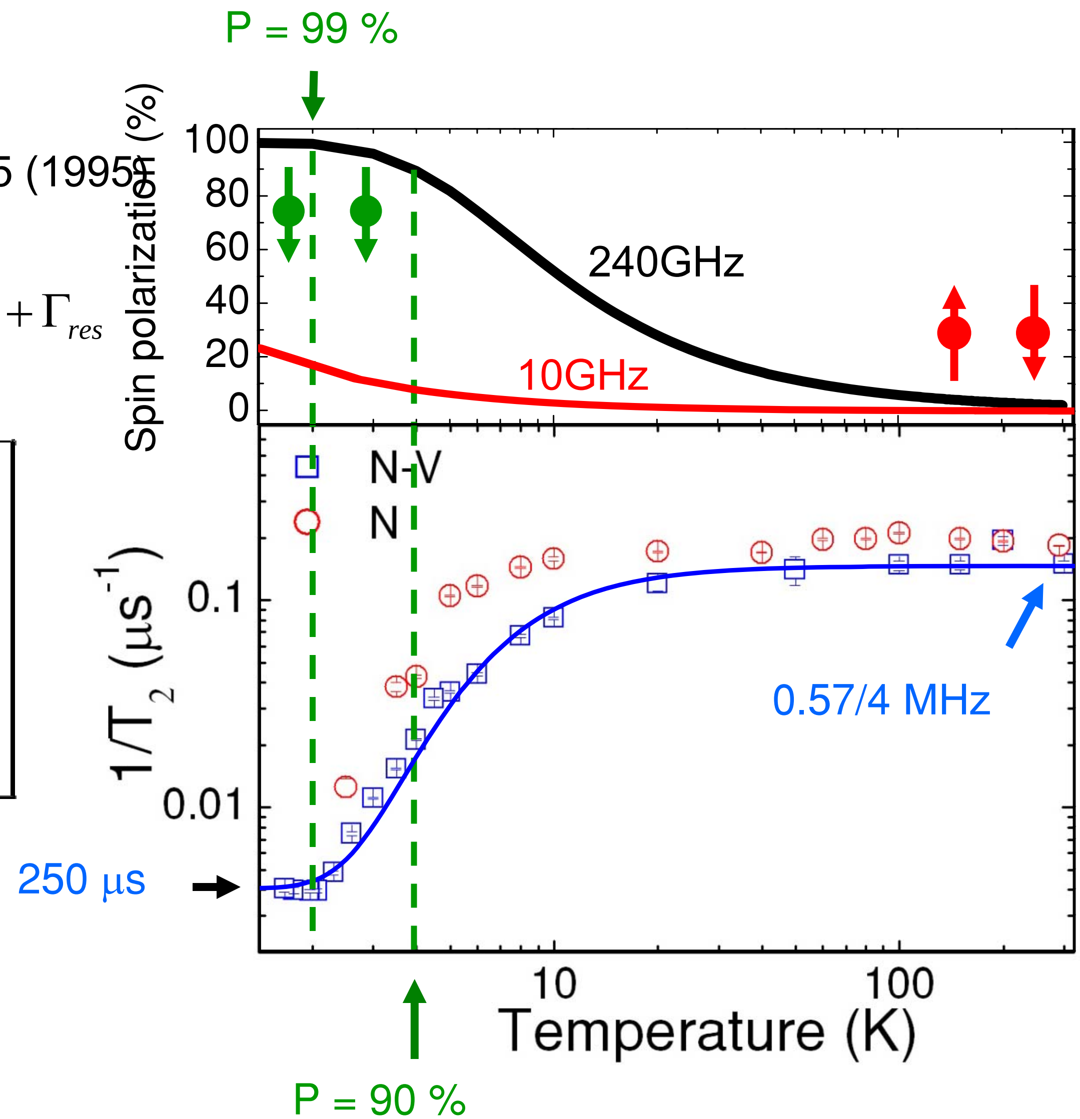
- N spin flip-flop process

C. Kutter *et al.*, *Phys. Rev. Lett.* **74**, 2925 (1995)

$$\frac{1}{T_2} = CP_{\uparrow}P_{\downarrow} + \Gamma_{res} = \frac{C}{(1+e^{T_{Ze}/T})(1+e^{-T_{Ze}/T})} + \Gamma_{res}$$

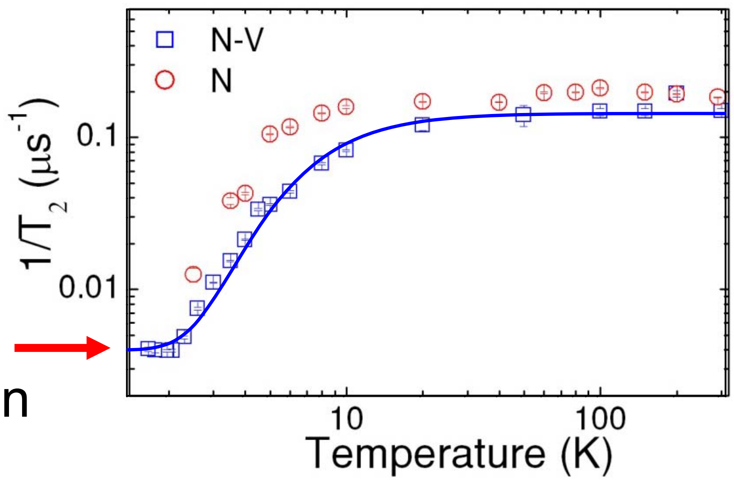


- C=0.57 MHz
- $1/\Gamma_{res}$ = 250 μs



^{13}C nuclear spin bath fluctuations

$$\frac{1}{T_2} \propto P_\uparrow P_\downarrow + \Gamma_{res} = \frac{C}{(1+e^{T_{Ze}/T})(1+e^{-T_{Ze}/T})} + \Gamma_{res}$$



- $1/\Gamma_{res}$ = 250 μs: Temperature independent relaxation rate

- Decoherence time caused by ^{13}C nuclear spin flip-flop process

$$\frac{1}{T_2} = 0.49\sqrt{\frac{\gamma_e}{\gamma_n}}\frac{\Delta\omega_{nn}}{[I(I+1)]^{1/4}} \sim \gamma_e^{1/2}\gamma_n^{3/2} N[I(I+1)]^{1/4}$$

where $\Delta\omega_{nn}$ is NMR linewidth, N is the number of nuclear per volume.

(I. M. Brown, Time domain electron spin resonance, p195, Wiley (1979).
A. Schweiger and G. Jeschke, Oxford university press (2001)).

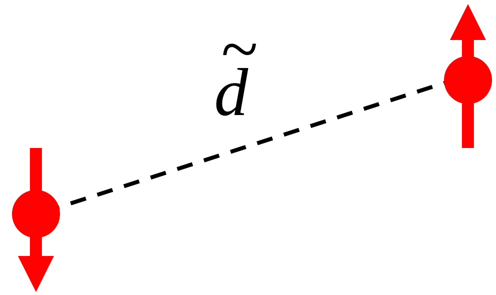
- T_2 ~ 380 μs for ^{13}C nuclear spin bath fluctuations

S. Takahashi *et al.*, *Phys. Rev. Lett.* **101**, 047601 (2008)

Dipole-dipole interaction

$$\frac{1}{T_2} \propto P_{\uparrow}P_{\downarrow} + \Gamma_{res} = \frac{C}{(1+e^{T_{Ze}/T})(1+e^{-T_{Ze}/T})} + \Gamma_{res}$$

Average distance



- C=0.57 MHz

- Dipole-dipole interaction energy

$$C = \frac{U_d}{h} \cong \left\langle \frac{\mu_0}{4\pi} \frac{\mathbf{m_1} \cdot \mathbf{m_2} - 3(\mathbf{n} \cdot \mathbf{m_1})(\mathbf{n} \cdot \mathbf{m_2})}{\tilde{d}^3} \right\rangle / h$$

- $\tilde{d}$ = 2.8 nm → N contents ~ 25 ppm
- The sample crystal = 10 ~ 100 ppm

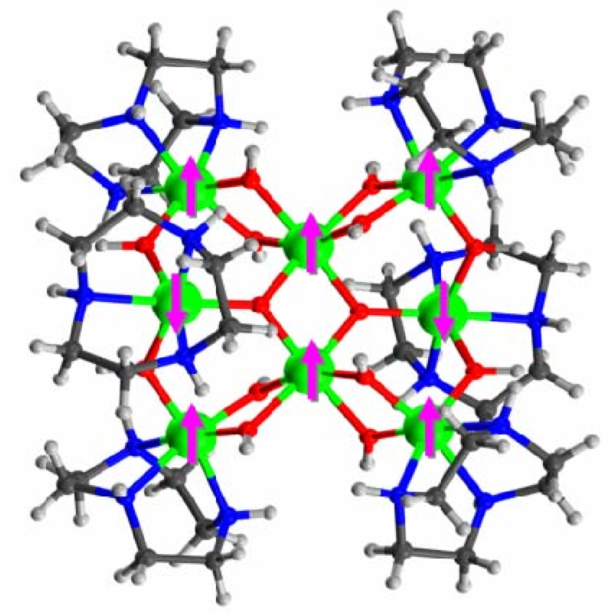
- **Potentially useful for T_2-based distance measurement**

Outline

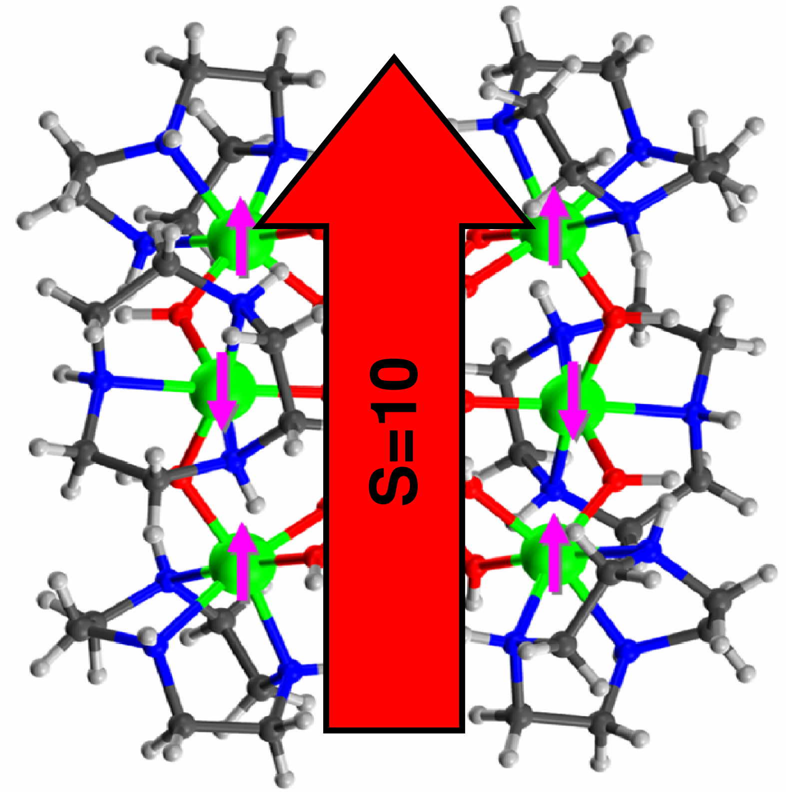
1. Quenching spin decoherence of NV centers in diamond

2. T_2 based distance measurement

3. Spin decoherence of Fe8 single-molecule magnets



Fe8 single-molecule magnets



● : Fe^{3+} (S=5/2) ● : O ● : N

● : C ● : H

K. Wieghardt *et al*., G. Angew. Chem., Int. Ed. Engl. 23, 77 (1984).

$[Fe_8O_2(OH)_{12}(C_6H_{15}N_3)_6]Br_7(H_2O)Br \cdot 8H_2O$

- High-spin molecular magnets made by metal-ion clusters
- Chemically fabricated nano-scale quantum objects
- S=10 Fe_8 SMM
  Total spin = (6-2)×5/2=10
- Weakly interacts with each other, ensemble properties express themselves as a pseudo-giant single spin.

Spin decoherence of SMMs

- Spin decoherence of SMMs *is* poorly understood because T_2s in most SMMs are too short to measure.

- There are some observations of spin echo from highly "*diluted*" molecular magnets.

  Cr_7Ni: A. Ardavan *et al.*, *Phys. Rev. Lett.* **98**, 057201 (2007)

  V_{15}: S. Bertaina *et al.*, *Nature* **453**, 203 (2008)

  Fe_4: C. Schlegel *et al.*, *Phys. Rev. Lett.* **101**, 147203 (2008)

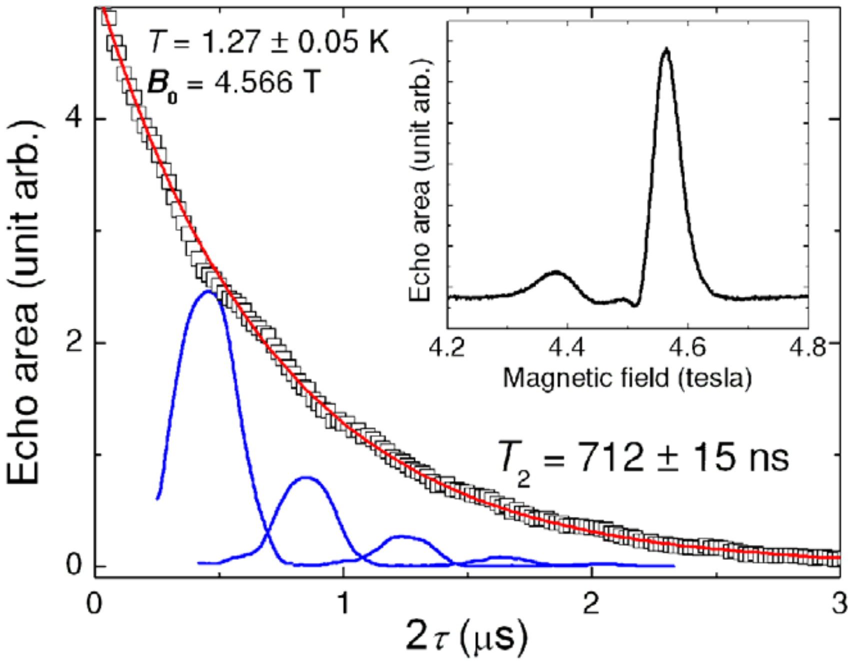
  Fe_7: Z. Wang et al., to be published.

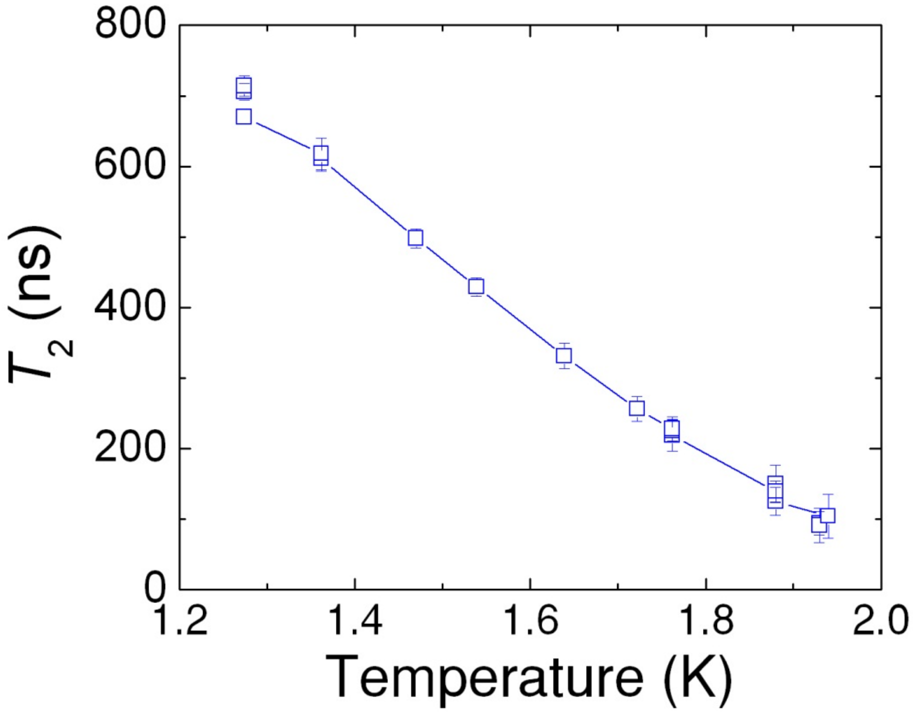
- Fluctuations of SMM spin bath can also be reduced with high-field EPR.

First spin echo measurement

- Strong temperature dependence indicates Fe_8 spin bath fluctuations.

T_2 measurement





S=10 Fe_8 spin bath fluctuations

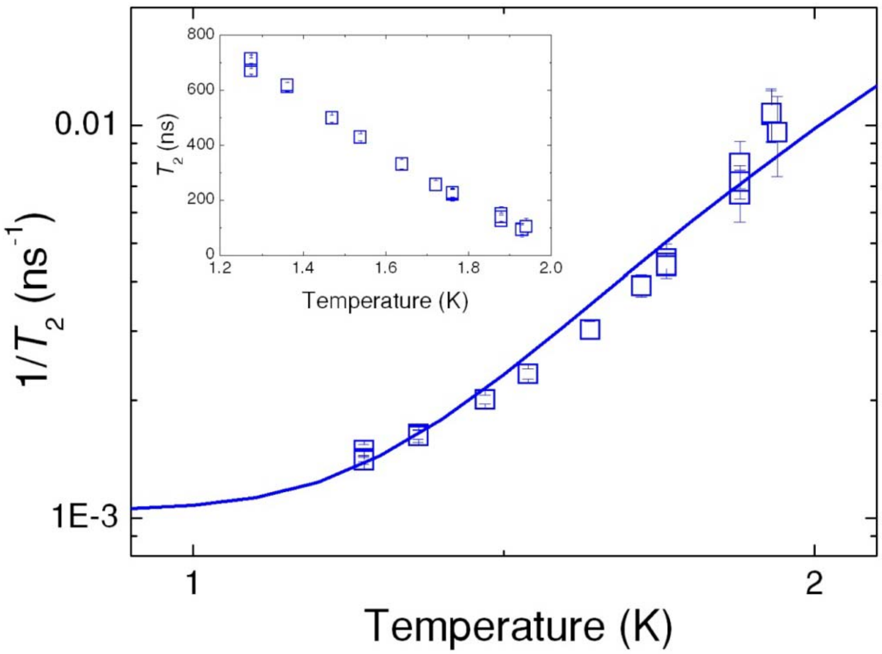
- S=10 spin flip-flop process:

$$\frac{1}{T_2} = A \sum_{m_S=-10}^{9} W(m_S)\, P_{m_S} P_{m_S+1} + \Gamma_{res}$$

$$P_{m_S} = \frac{e^{-\beta E(m_S)}}{Z}$$

$$W(m_S) = \left|\left\langle m_S+1, m_S \left| S_1^+ S_2^- \right| m_S, m_S+1 \right\rangle\right|^2$$

$$+ \left|\left\langle m_S, m_S+1 \left| S_1^- S_2^+ \right| m_S+1, m_S \right\rangle\right|^2$$



- Spin decoherence is significantly suppressed by spin polarization.

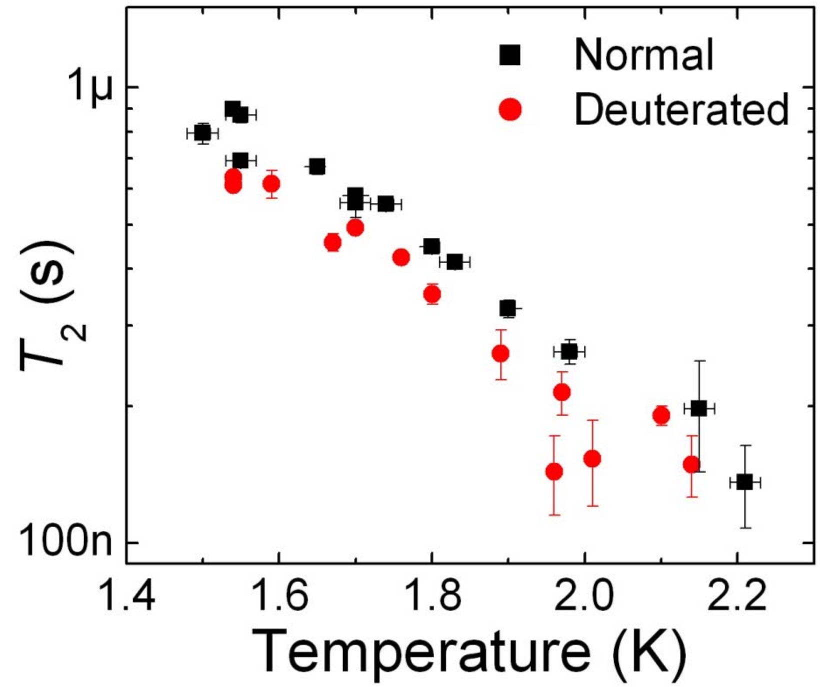
- Γ_{res} may be nuclear moments (^{1}H) and phonons.

S. Takahashi *et al.*, *Phys. Rev. Lett.* **102**, 174102 (2009)

No ^{1}H spin bath decoherence

- Deuterated Fe_8 SMM

- Deuterium has much smaller magnetic moments than ^{1}H.



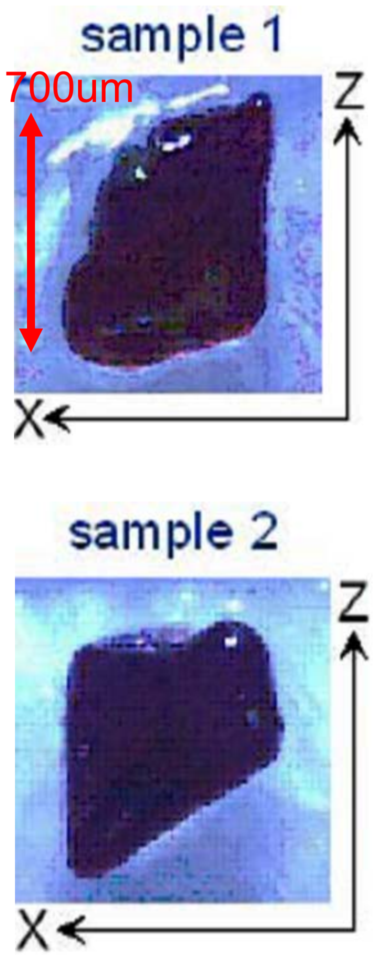
$[Fe_8O_2(OH)_{12}(C_6H_{15}N_3)_6]Br_7(H_2O)Br \cdot 8H_2O$

^{2}D ^{2}D ^{2}D

- Temperature dependence of T_2 is about same.

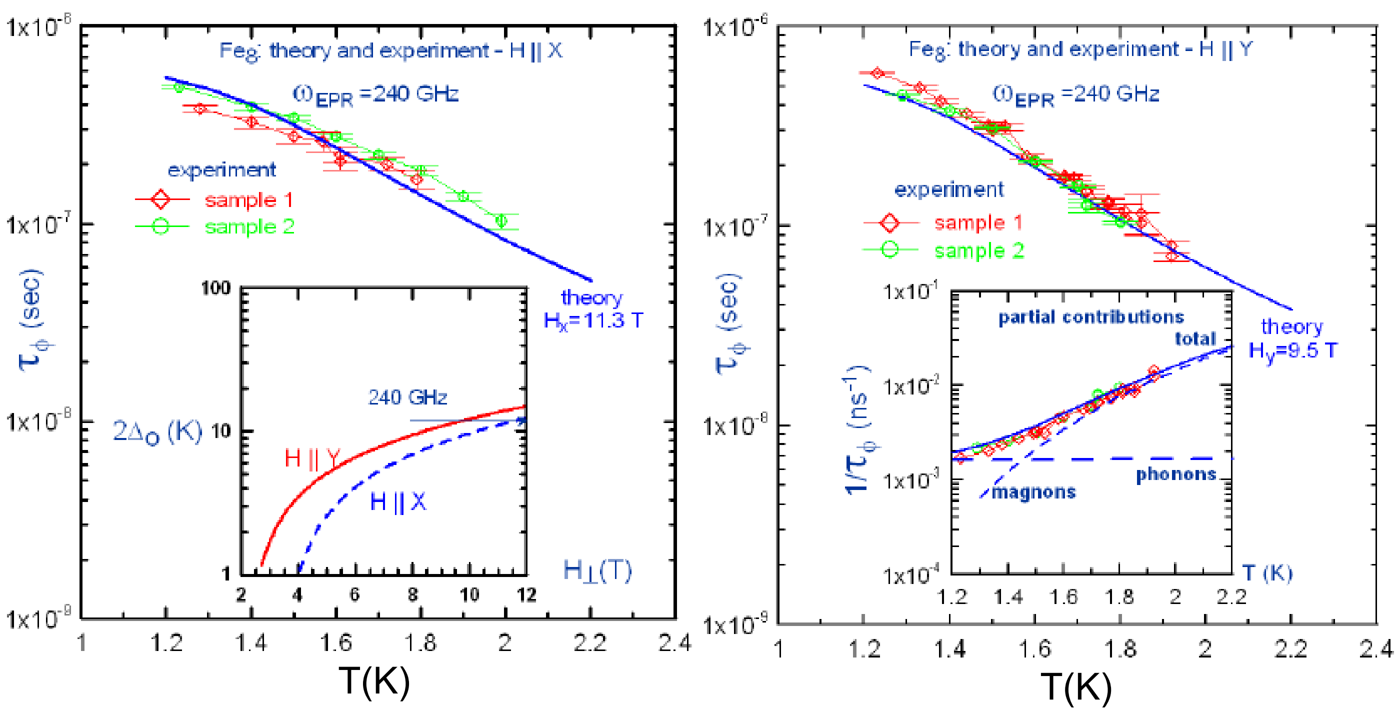
S. T. and C. Beedle *et al.*, to be published

Theory - experiment



A. Morello, I. Tupitsyn and P. Stamp, *Phys. Rev. Lett.* **97**, 207206 (2006)

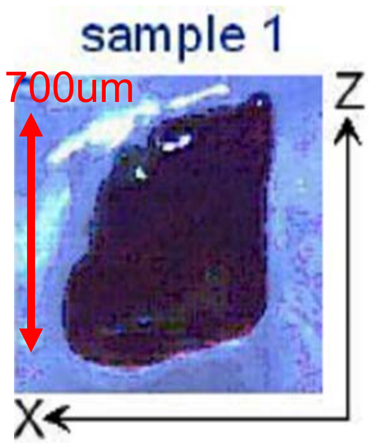
Spin decoherece: $1/\tau_\phi = \Gamma_{d\text{-}d} + \Gamma_{ph} + \Gamma_{nuc}$

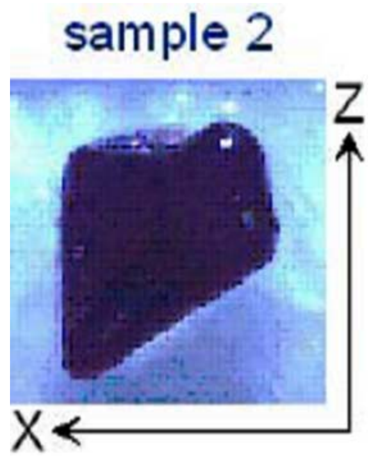


No fitting parameters!

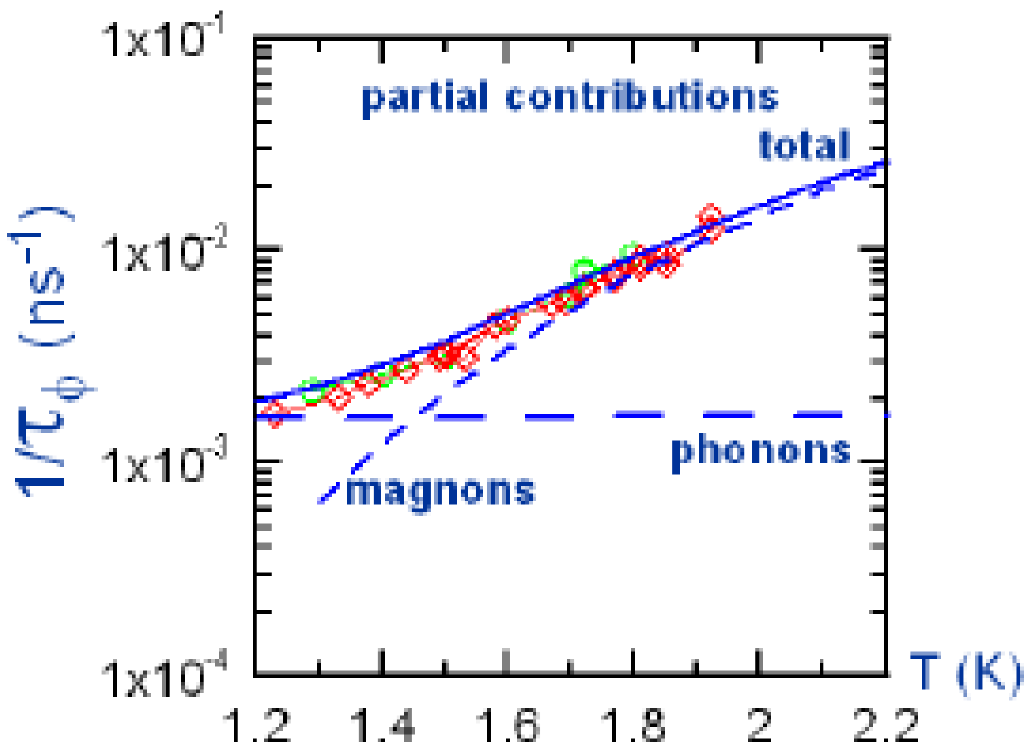
Theory - experiment

A. Morello, I. Tupitsyn and P. Stamp, *Phys. Rev. Lett.* **97**, 207206 (2006)





Spin decoherece: $1/\tau_\phi = \Gamma_{d\text{-}d} + \Gamma_{ph} + \Gamma_{nuc}$



S.T., I.T. and P.S. *et al.*, manuscript in preparation

Summary

- Electron spin bath decoherece can be quenched by high fields and low temperature.
- HF EPR can access to other decoherence (nuclear spin bath and phonons)
- HF EPR can be used for distance measurement.

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