Decoherence in Solid State

superconducting circuits, quantum dots, magnetic molecules, quantum phase transitions

B. Barbara

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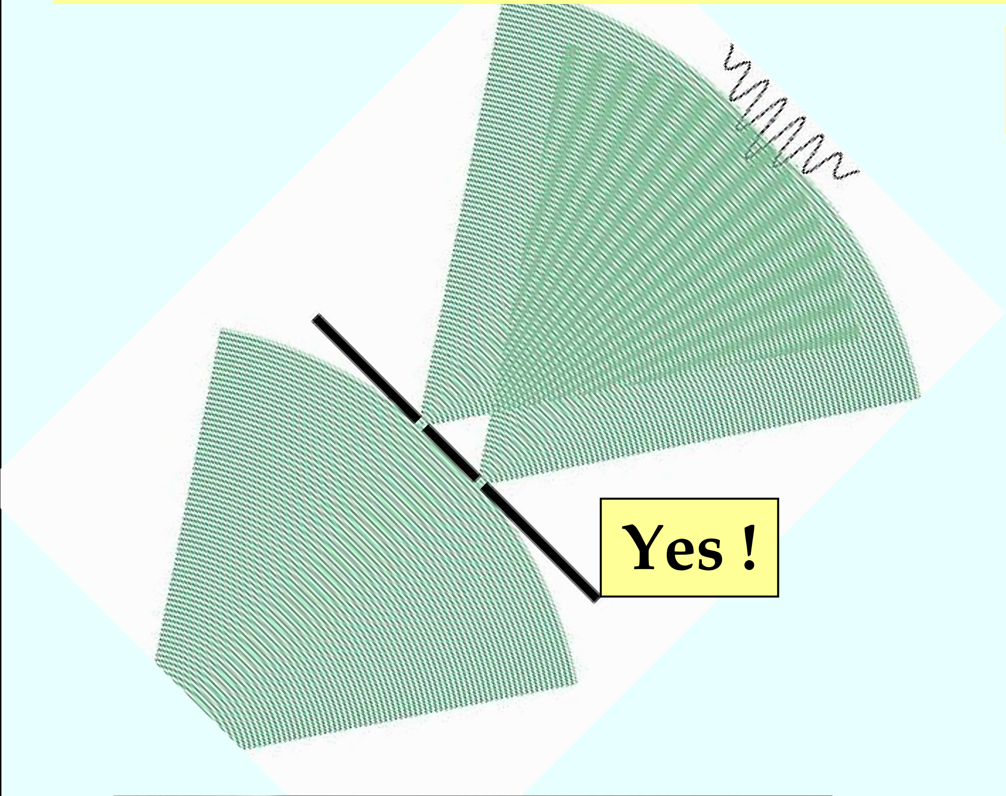
Large scale diffraction, interferences

shape their environmement

Sao Martinho de Porto (Portugal)

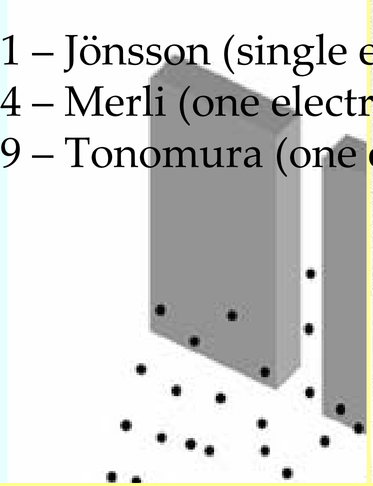
« Waves » of photons, electrons,…

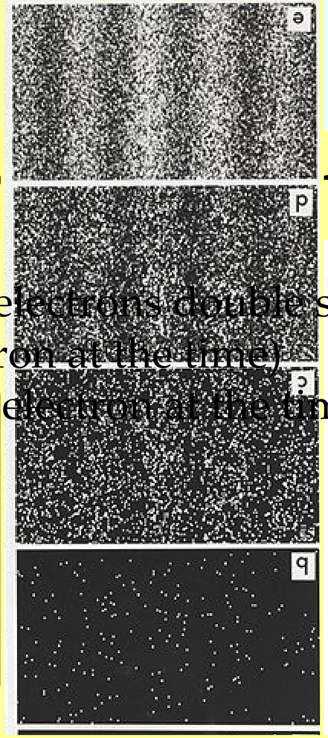


Yes !

Single photor ns..

1961 – Jönsson (single electrons double slit)
1974 – Merli (one electron at the time)
1989 – Tonomura (one electron at the time)

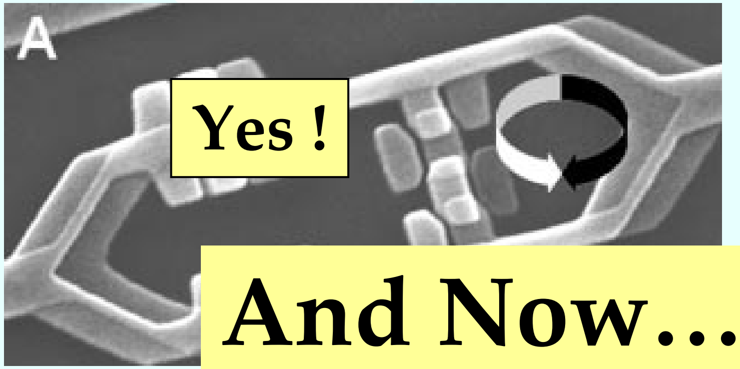




The roots of decoherence:

1970 – Zeh
1987 – Simonius
1980 – Zurek… Giulini, Schlosshauer
1981 – Leggett, Caldeira (dissipation)
1996 – Stamp, Prokof’ev (spin-bath)

« Decoherence »

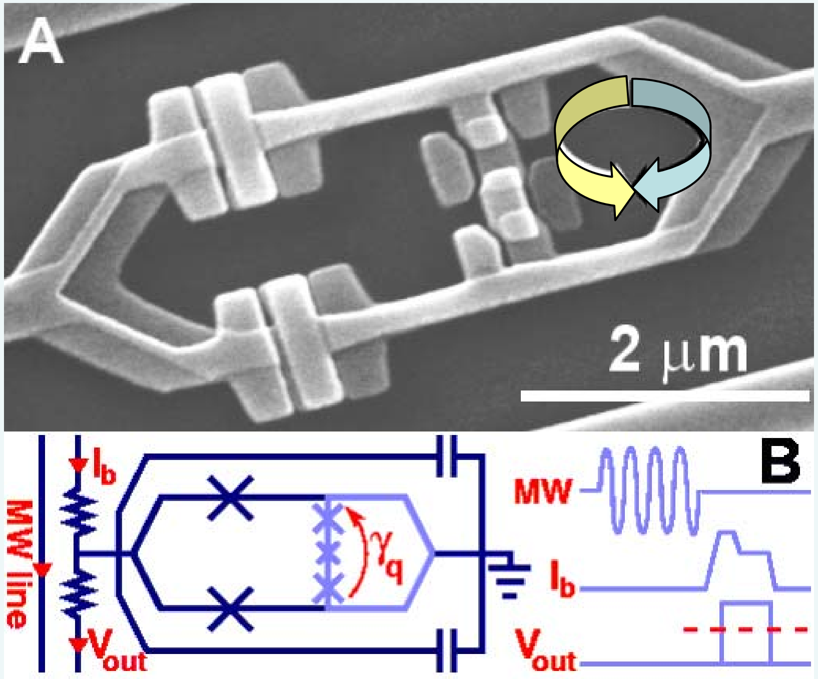


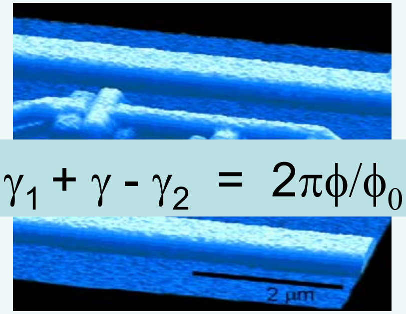
Yes !

And Now…

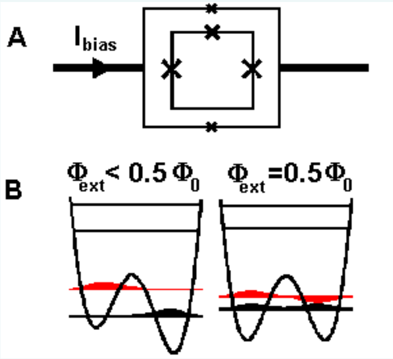
Coherent control of a superconducting flux qubit

I. Chiorescu, Y. Nakamura, J.P.M Harmans, J.E. Mooij, Delft (2003)



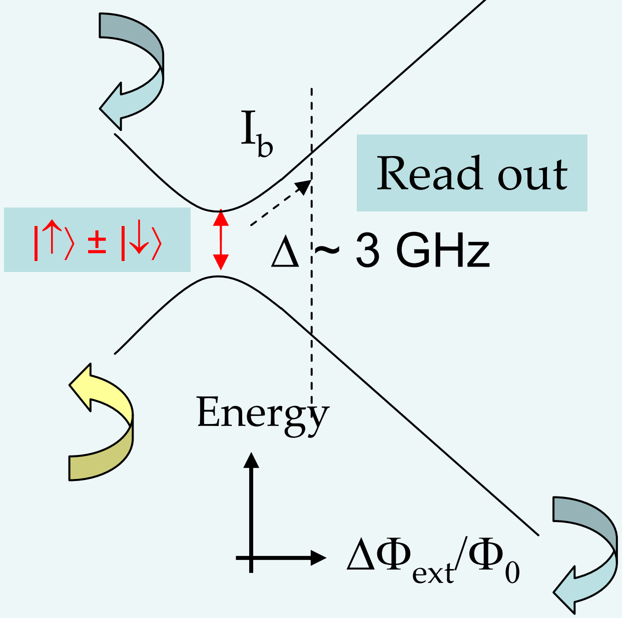


$\gamma_1 + \gamma - \gamma_2 = 2\pi\phi/\phi_0$



$E_J >> E_c$

Insensitive to charge fluctuations



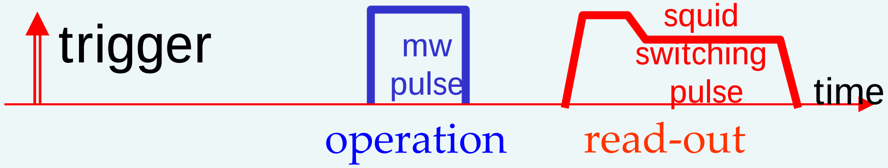
Read out

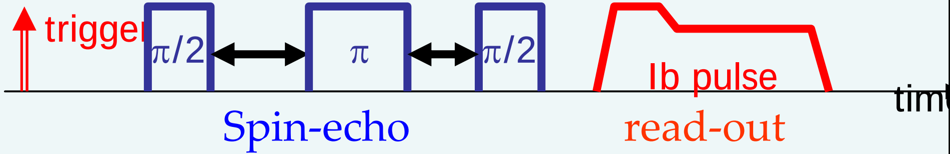
$|\uparrow\rangle \pm |\downarrow\rangle$

Δ ~ 3 GHz

This first experimental coherent manipulation of flux qubits follows the line opened by Nakamura (NEC, Tsukuba), and the Saclay Quantronics group (Devoret, Esteve)

Rabi oscillations of the flux qubit





$F_{\text{Larmor}} = 6.6$ GHz

Relaxation $T_1 \approx 1\ \mu s$

Rabi decay $T_R \approx 150$ ns

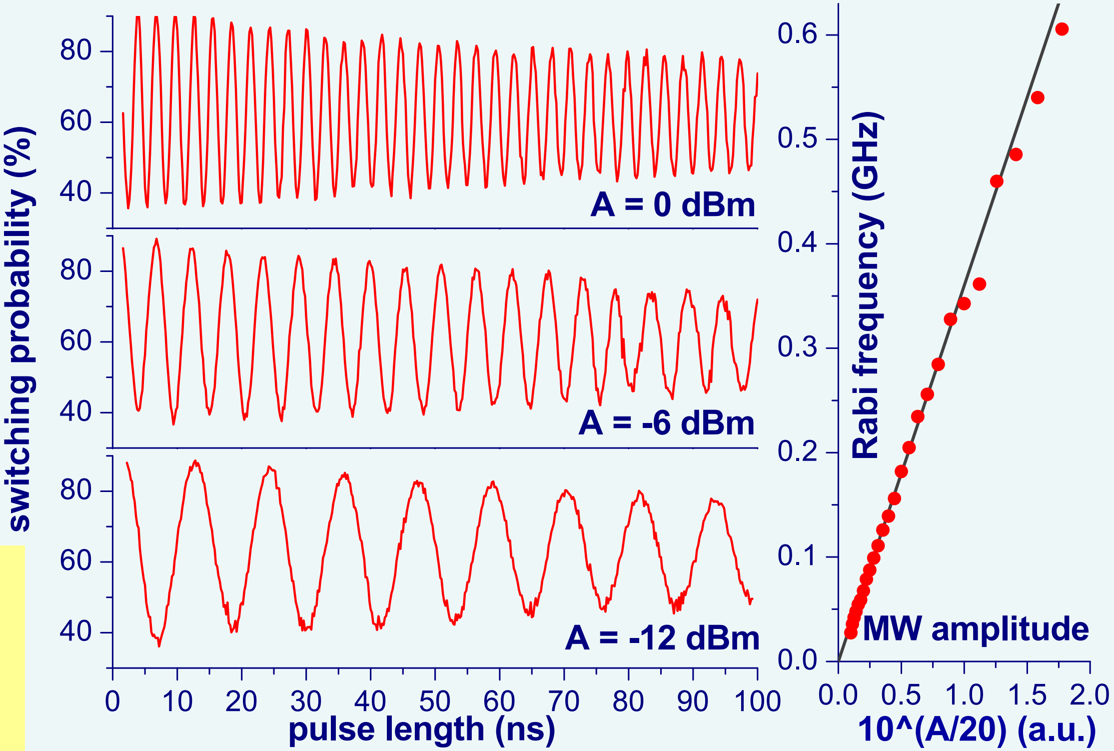
Ramsey $T_{Rm} \approx 20$ ns

Spin-echo $T_2 \approx 30$ ns

Many oscillations (large Ω_R)

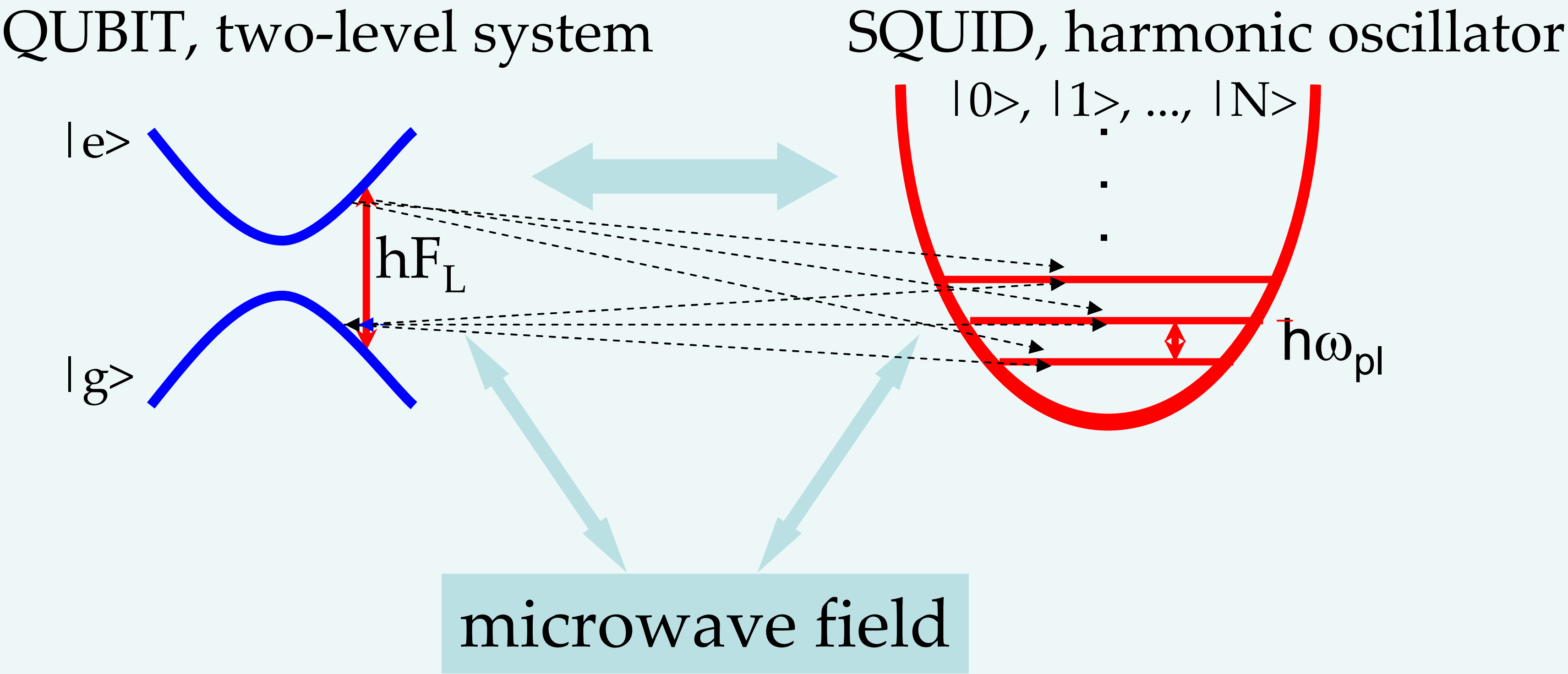
Free and driven coherence limited by 1/f (noise) >> 10 MHz (Difficult to avoid and to predict)

Read-out fidelity ~ 50%



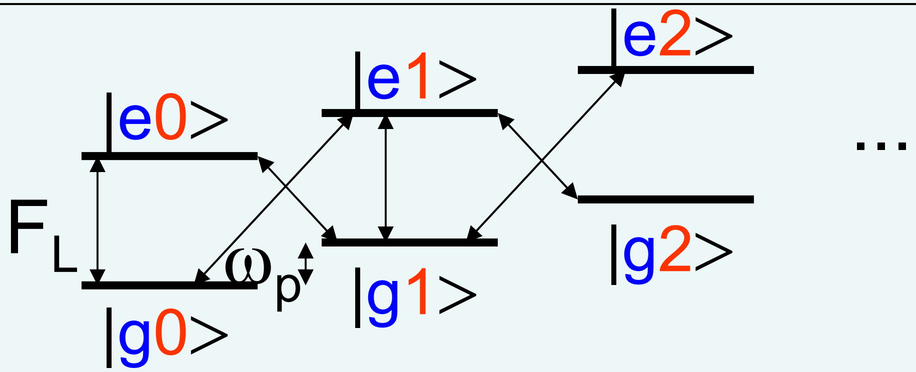
I. Chiorescu, Y. Nakamura, C.J.P.M. Harmans, J.E. Mooij, *Science*, **299**, 1869 (2003)

Entanglement between flux qubit and measuring tool (SQUID)



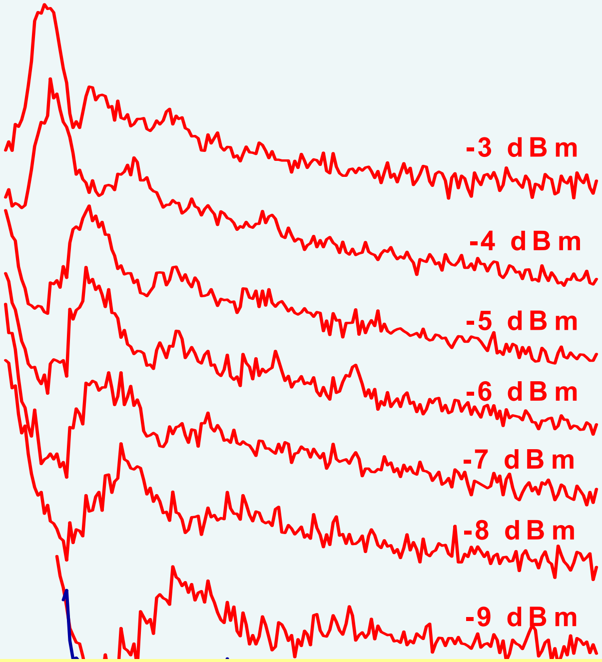
QUBIT, two-level system

SQUID, harmonic oscillator



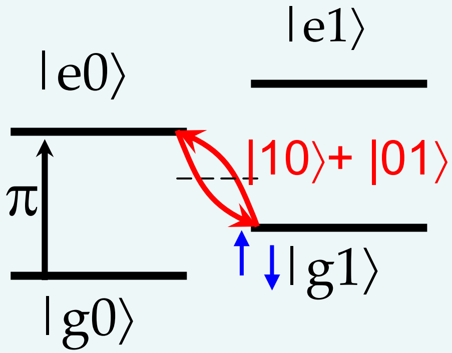
Driven Rabi oscillations of the entangled system

I. Chiorescu, P. Bertet, K. Semba, Y. Nakamura, C.J.P.M. Harmans, J.E. Mooij , *Nature* **451**, 139 (2004).



π pulse: $|g0\rangle \Rightarrow |e0\rangle$

Microwave: $|e0\rangle \Leftrightarrow |g1\rangle$



Oscillations between entangled states:

Strong decoherence (T_R ~ 3ns) by SQUID relaxation (level broadening, T_1~ 6 ns)

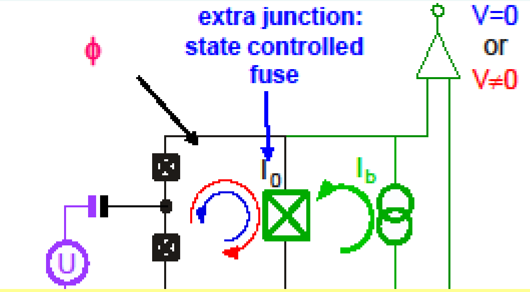
Should affect qubit Rabi oscillations (g0 ⟷ e0)

« **Quantronium** » = charge qubit and flux read-out
(and inversely)

The quantronium: « a major roadblock dissolved ? »

A.J. Leggett, Science, 3 May, 2002.

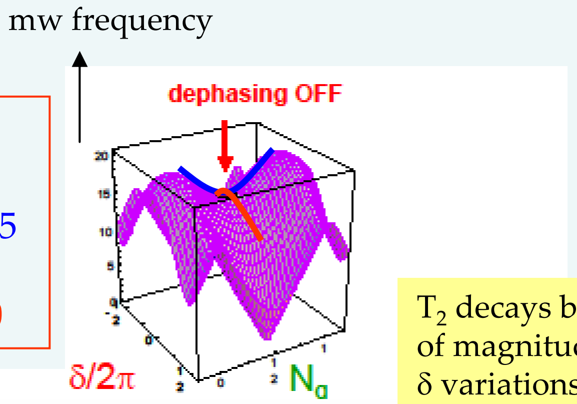
$E_J \approx E_{CP}$ → neither island charge nor phase is a good quantum number



Immunity

Flux qubit: Ng = 0.5

Charge qubit: $\phi = 0$



T_2 decays by 2 orders of magnitude for N_g or δ variations of 0.1

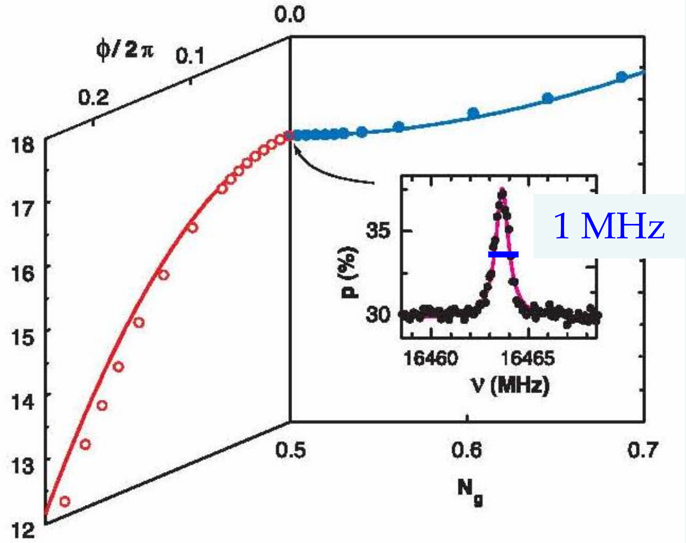
Better coherence (before $T_{Rm} \approx 20 \rightarrow 500$ ns)

Large oscillation frequencies → large Q_ϕ

Still, 2nd order decoherence, "electrical" 1/f noise

Read-out fidelity, still poor.

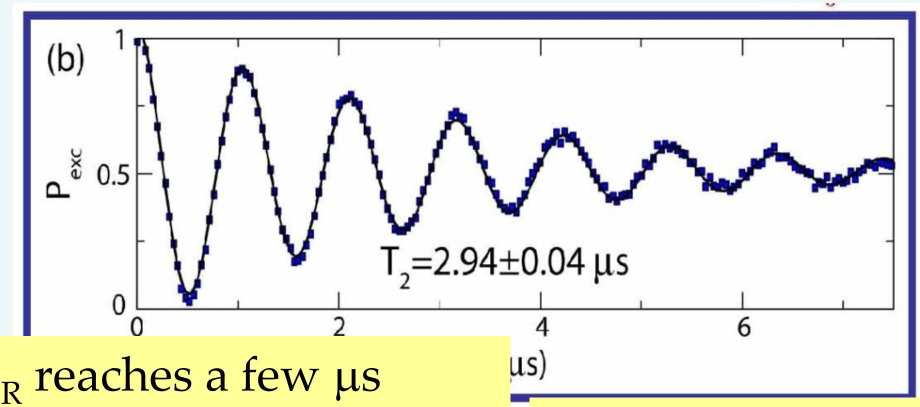
Difficult to do better → use of mw cavity



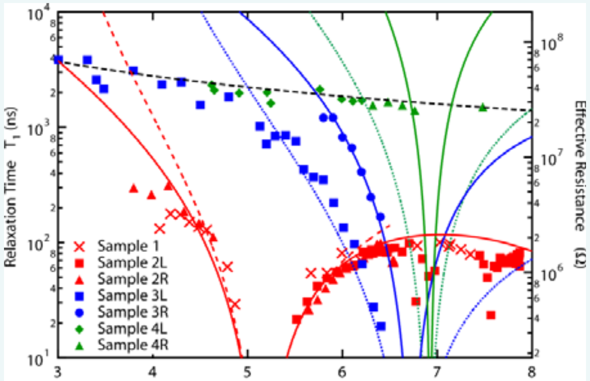
D. Vion, A. Aassime, A. Cottet, P. Joyez, H. Pothier, C. Urbina, D. Esteve, M. H. Devoret, Science, 3 May, 2002.

Last steps: qubit embedded in a 1D microwave cavity

High degree of control of e-m environment, High fidelity read-out… low power



T_R reaches a few μs
Fidelity approaches 95%
(even with single shots)
Factor of merit : limited



In all cases, coherence is limited by T_1

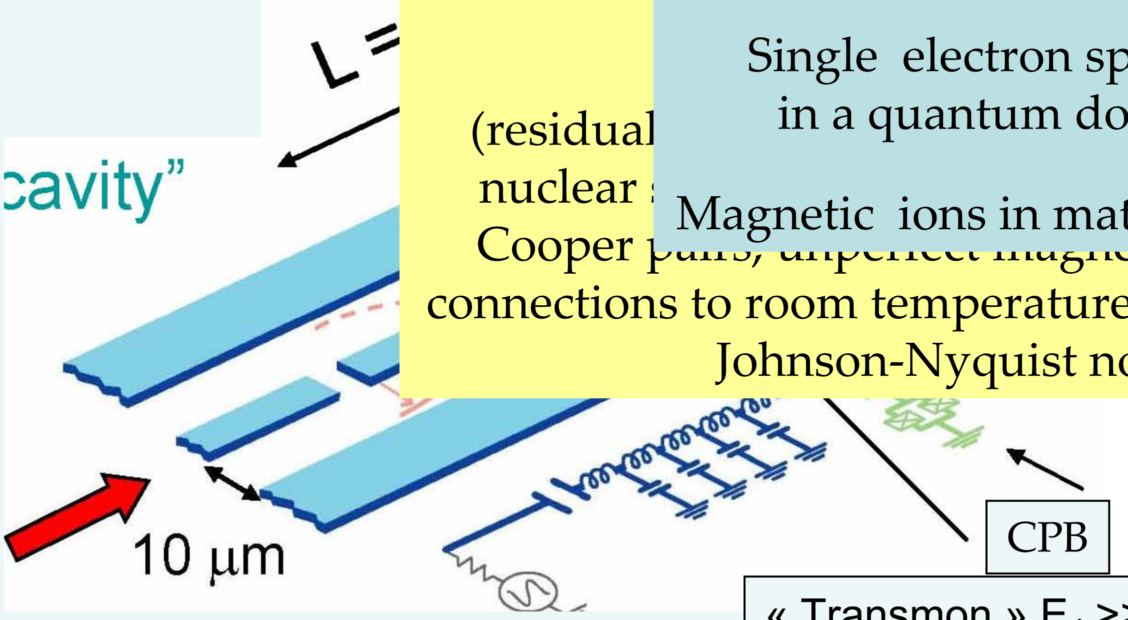
and … oseconds

(residual … s, impurities, nuclear … excitation of Cooper pairs, unperfect magnetic shielding, wire connections to room temperature measurement parts, Johnson-Nyquist noise …)

What about spins ?

Single electron spin
in a quantum dot

Magnetic ions in matrices



« Transmon » $E_J >> E_C$

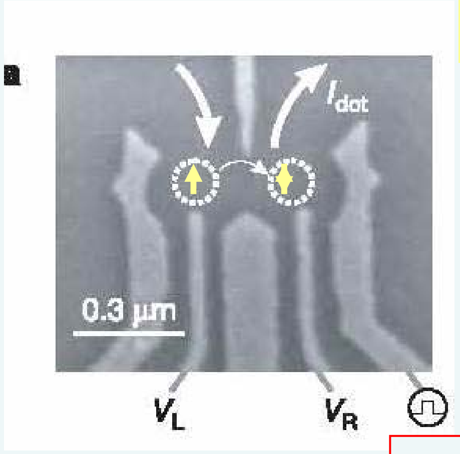
insensitive to charge noise,
see also Nakamura,(1999)

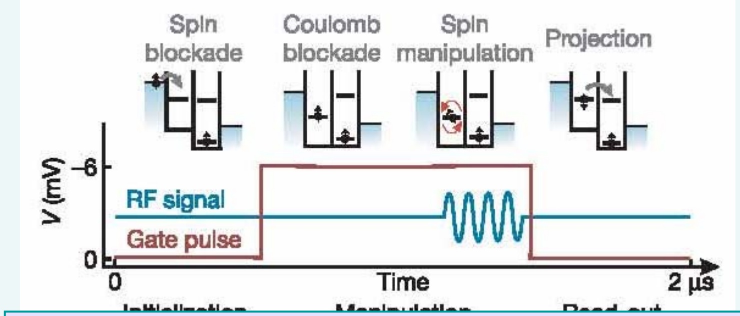
Blais et al, PRA, 2004
Walfraff et al, nature, 2004
…
Mallet et al, nature Phys, 2009

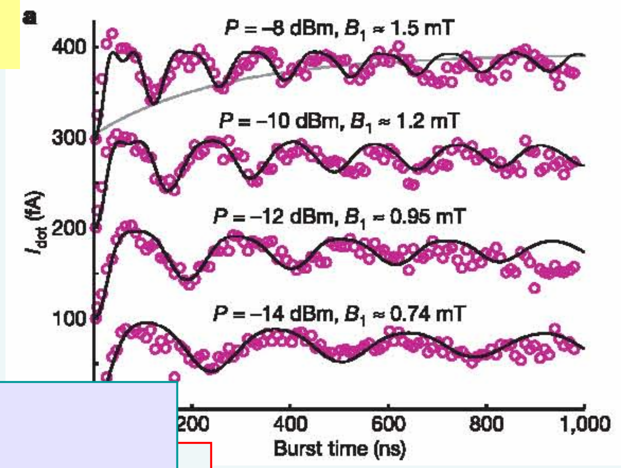
Driven oscillations of a single electron spin in a quantum dot

F. Koppens, C. Buizert, K.J. Tielrooj, I.T. Vink, K.C. Nowack, T. Meunier, L.P. Kowenhoven, L. Vandersypen
Nature, 17 Aug., 2006

Singlet and triplet states entangle with nuclear spin states ($\Delta \approx \sigma$)







T_{R}

$T_{S\text{-}B} \gg T_{ReadOut}$

$T_{S\text{-}B} \ll T_{Int}$

Nuclear S-B is fro
during each measur

Distribution of Larmor f

Coherence limitations

Slow N-S fluctuations affect free coherence

Fast N-S fluctuations ($B_{N\text{-}S} \sim B_{mw}$) affect spin manipulations → low fidelity

↓

« Disentangle » E-N spins
Suppress NS (graphen , C-nanotubes…)

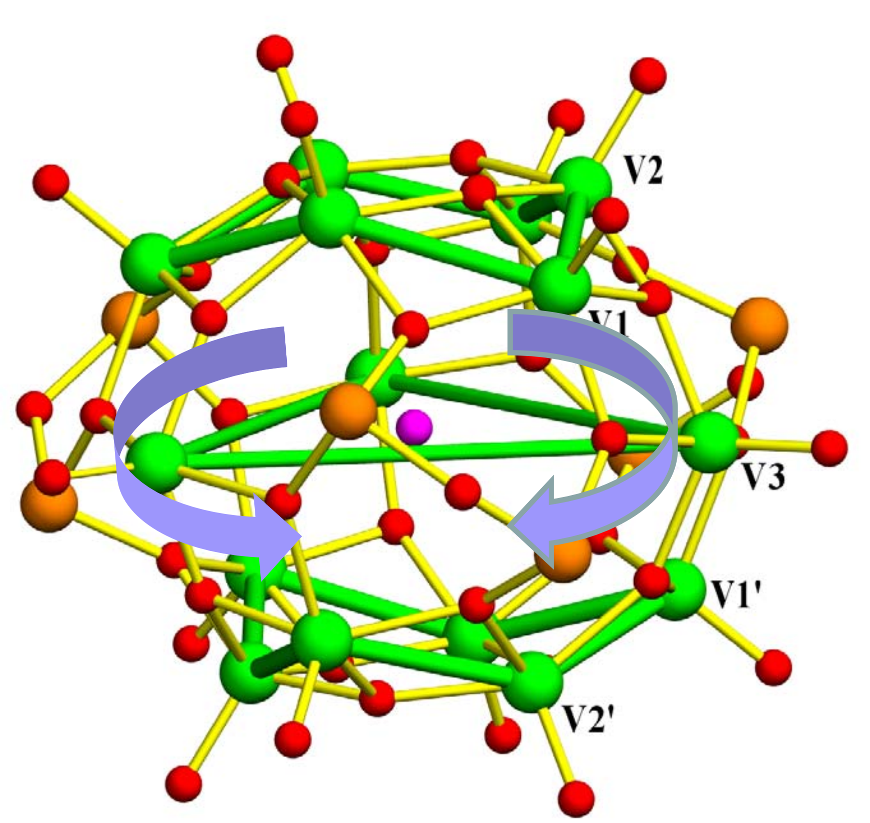
Average over
tributed frequencies

ctive additions, $T_2^* \sim h/\sigma$

Rabi oscillations (driven) and Spin-echo (suppression of slow NS fluctuations)
$T_R \approx T_{S\text{-}E} \approx 1\ \mu s$

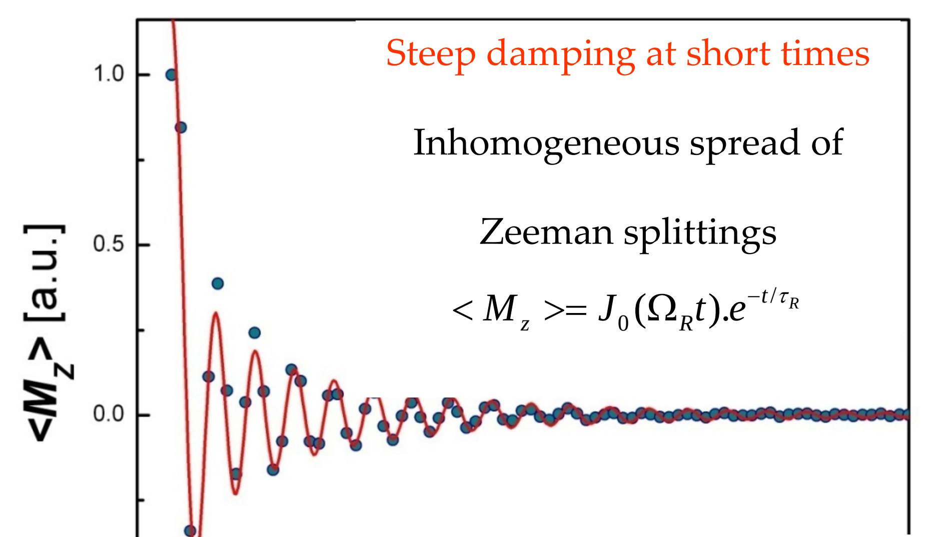
Magnetic ions in matrices

N-V centers in diamond, 3d (e.g. Mn), 4f (e.g. Er, Yb)
Molecular magnets



V_{15} : 15 spins 1/2, Hilbert space dimension $D = 2^{15} \sim 10^6$

Rabi oscillations in V_{15}

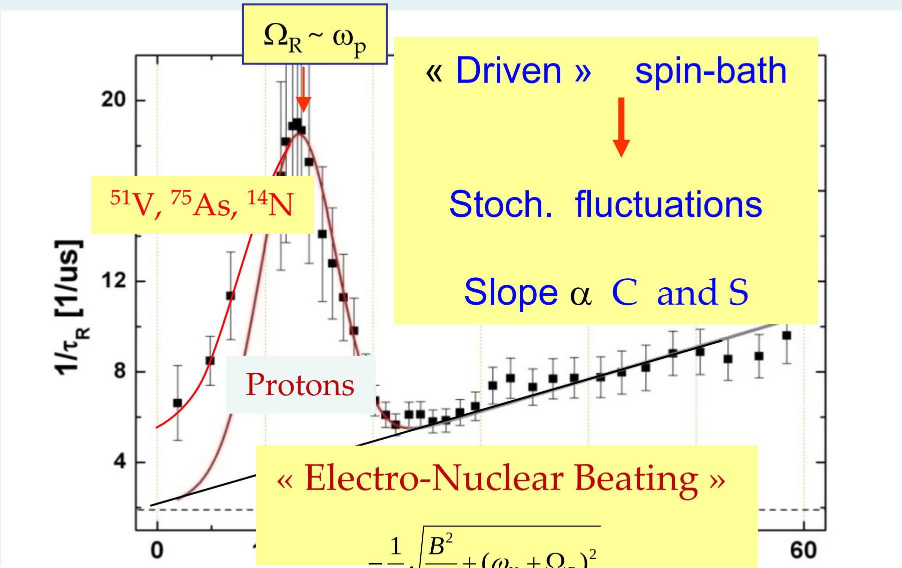


Steep damping at short times

Inhomogeneous spread of

Zeeman splittings

$$< M_z >= J_0(\Omega_R t).e^{-t/\tau_R}$$



$\Omega_R \sim \omega_p$

« Driven » spin-bath

Stoch. fluctuations

Slope α C and S

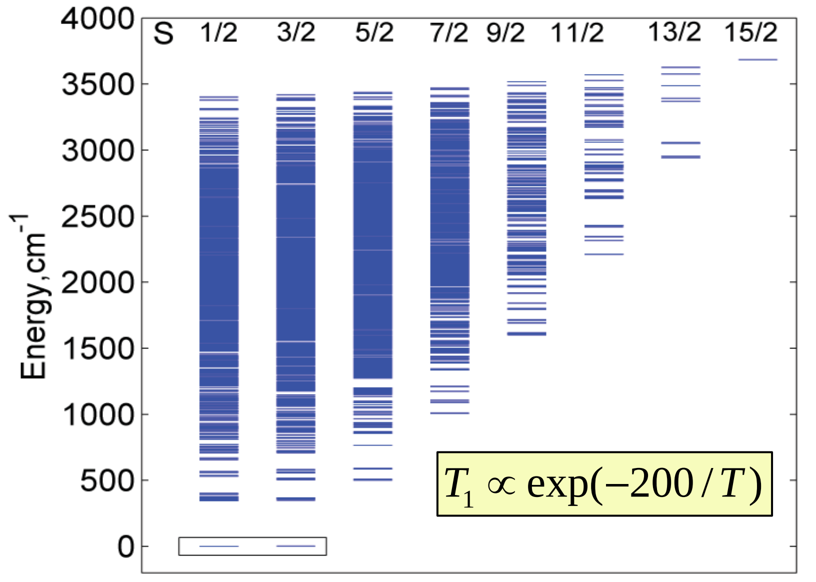
^{51}V, ^{75}As, ^{14}N

Protons

« Electro-Nuclear Beating »

$$-\frac{1}{}\sqrt{\frac{B^2}{4}+(\omega_N+\Omega_R)^2}$$

Similar to Modulated oscillations Single N-V centerSt Barbara



$$T_1 \propto \exp(-200/T)$$

Pairw

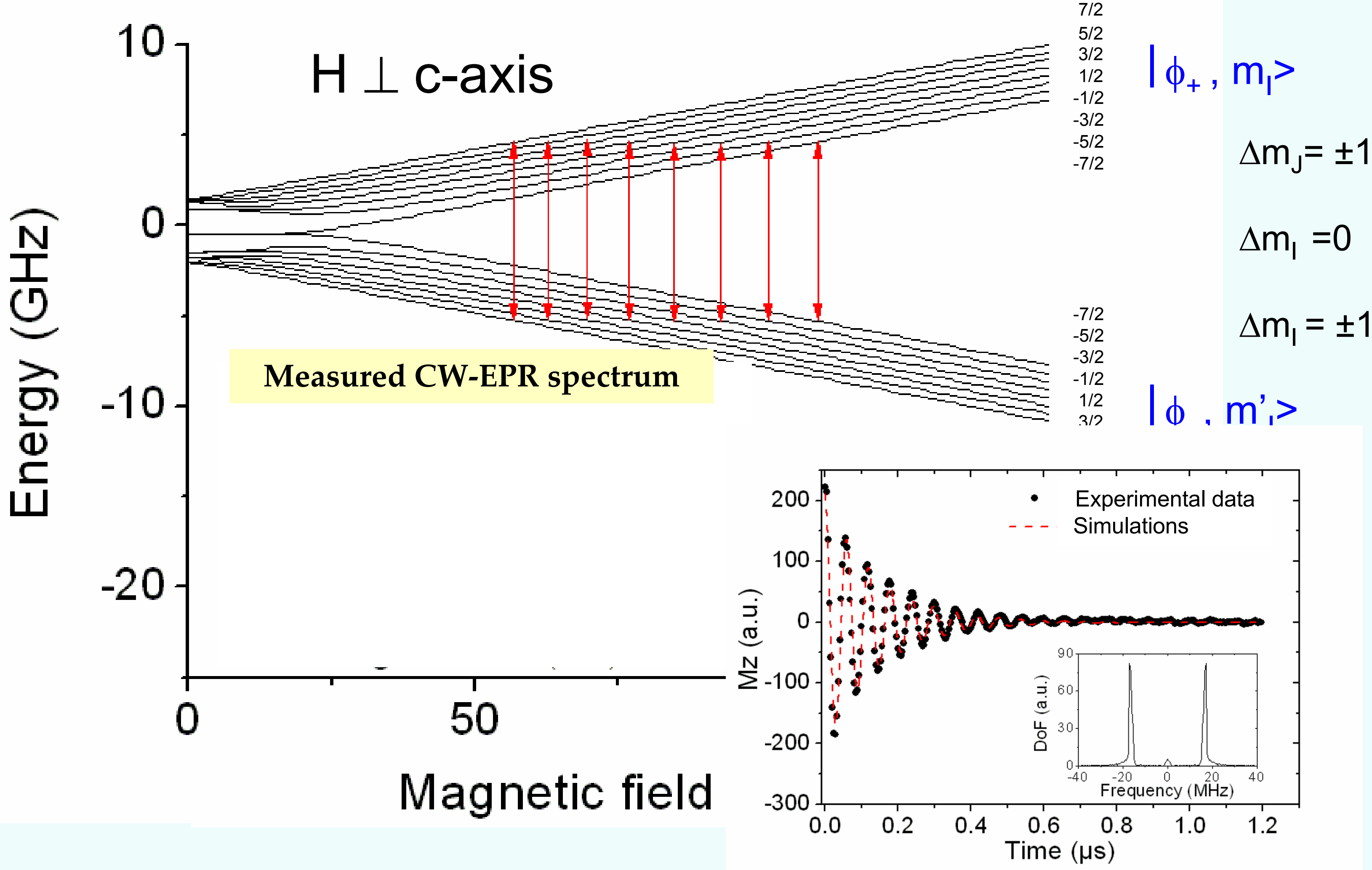
Quant

lings

odel

Rare-earth qubits (I=7/2 isotope)

CF ground-state + Hyperfine Interactions $^{167}Er^{3+}$: $CaWO_4$



$|\phi_+ , m_I>$

$\Delta m_J = \pm 1$

$\Delta m_I = 0$

$\Delta m_I = \pm 1$

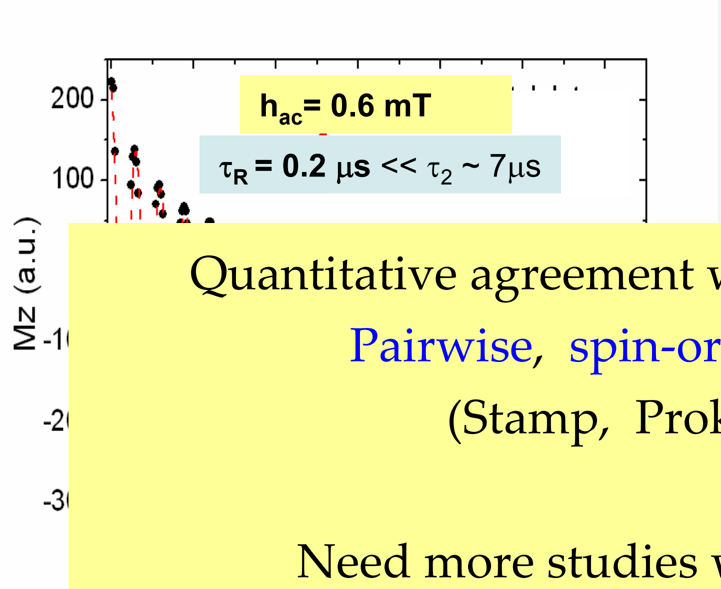
$|\phi$, $m'_I>$

Measured CW-EPR spectrum

Eight independent electro-nuclear transitions

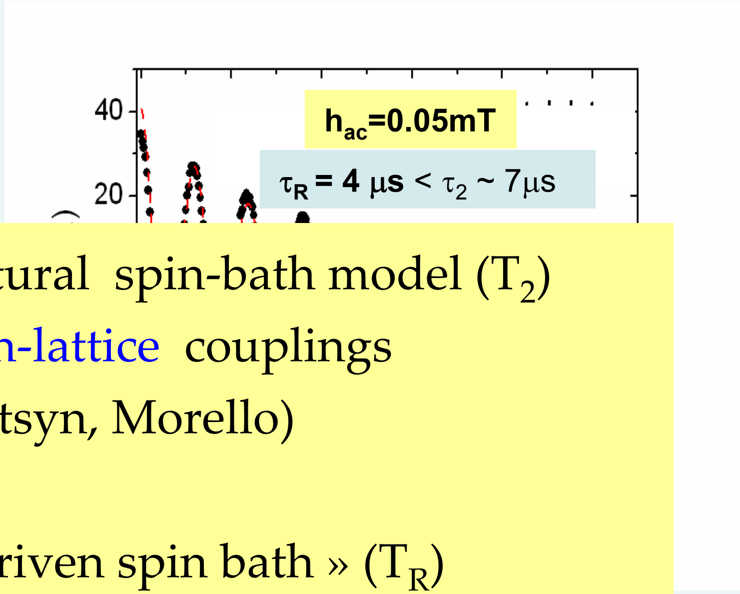
Nature nanotechnology (2007)

Damping of oscillations
Effect of the microwave power



h_{ac}= 0.6 mT

τ_R = 0.2 µs << τ_2 ~ 7µs



h_{ac}=0.05mT

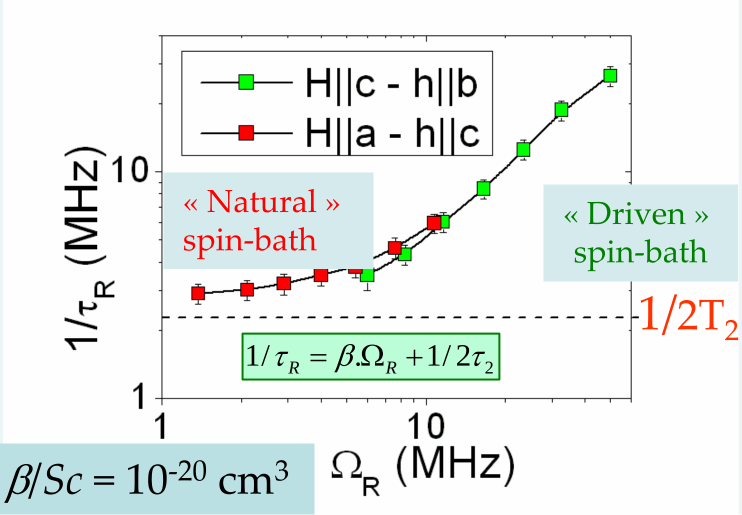
τ_R = 4 µs < τ_2 ~ 7µs

Quantitative agreement with the natural spin-bath model (T_2)

Pairwise, spin-orbit and spin-lattice couplings

(Stamp, Prokof'ev, Tupitsyn, Morello)

Need more studies with the « driven spin bath » (T_R)



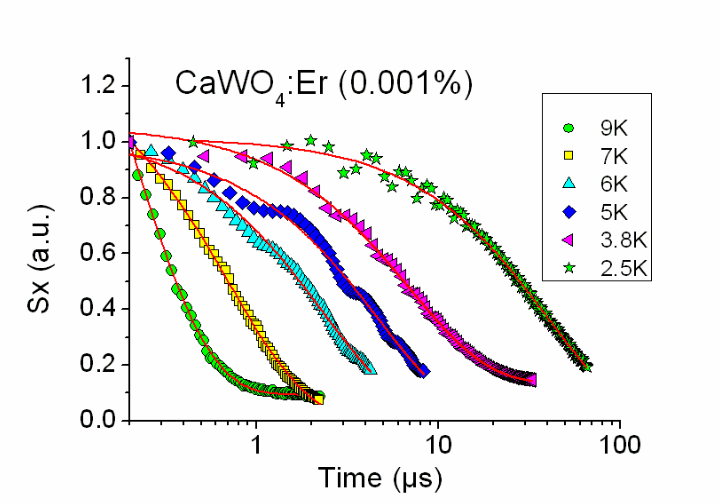
« Natural » spin-bath

« Driven » spin-bath

$1/2T_2$

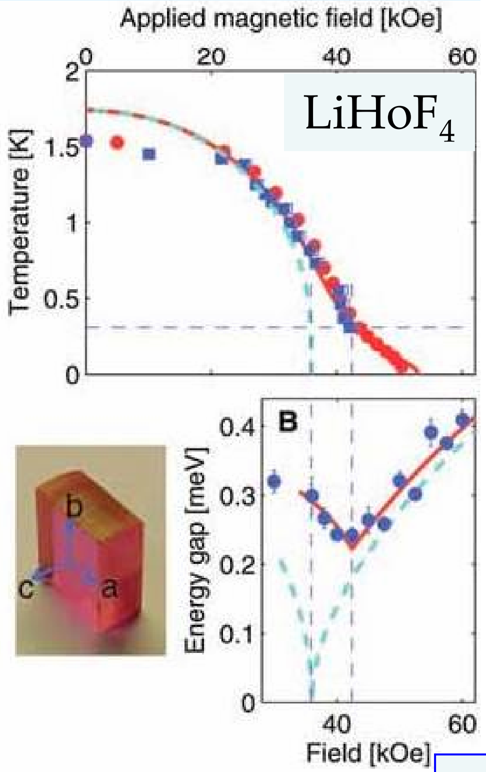
$1/\tau_R = \beta.\Omega_R + 1/2\tau_2$

$\beta_N = \beta/Sc = 10^{-20}$ cm^3

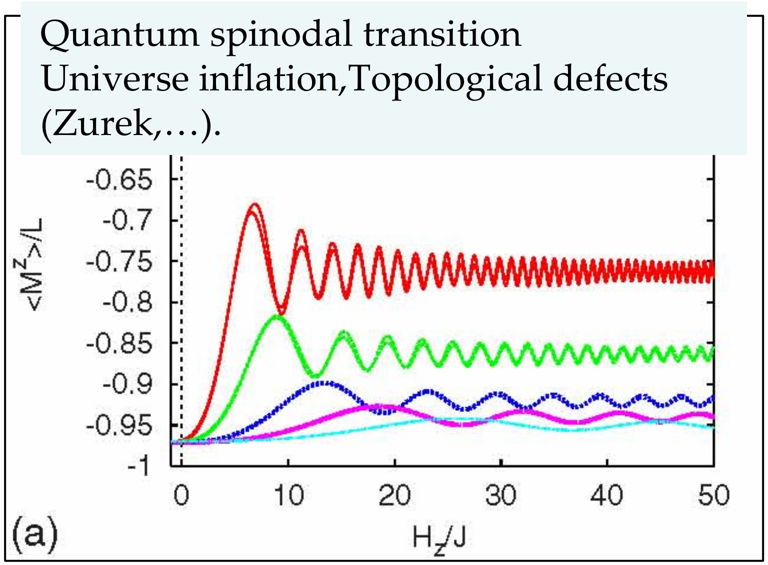


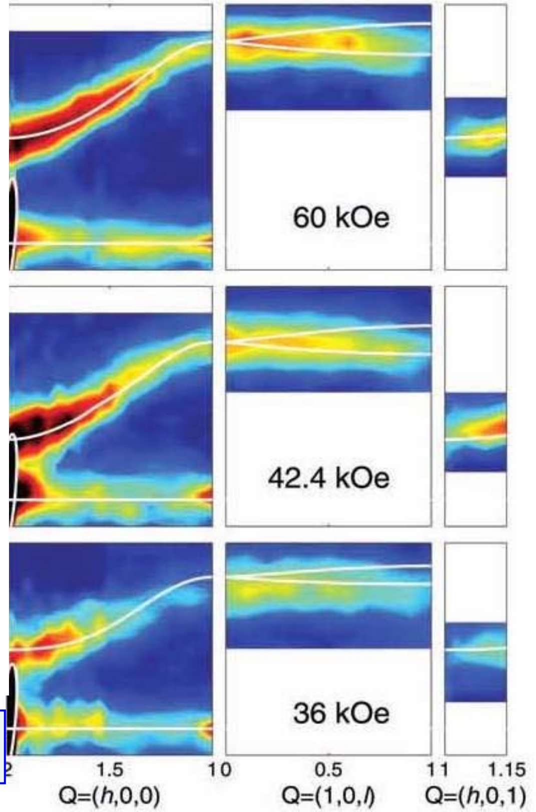
Quantum phase transition of a magnet in a spin-bath

H. Ronnov, R. Parthasarathy, J. Jansen, G. Aeppli, T. Rosenbaum, D. McMorrow, Science, 15 April 2003



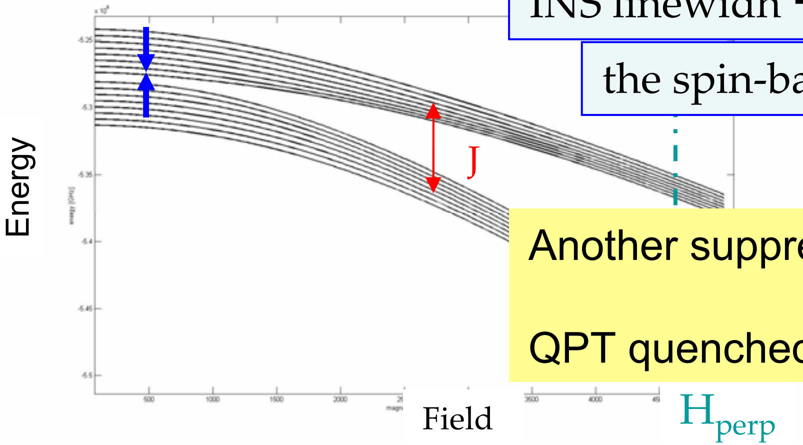
Quantum spinodal transition
Universe inflation,Topological defects
(Zurek,…).





INS linewidh → ξ_c ~ 1 interatomic distance.

the spin-bath suppresses the QPT



Another suppression mechanism:

QPT quenched by fast field sweepings (period, entanglement length)

THANK YOU

FOR YOUR ATTENTION !

Conclusions

Superconducting qubits: Important progess in coherence times and fidelity (95% for « single shot » measurements) due to wonderful experimental tricks and technical prowes
Important developments in nanosciences and nanotechnologies.
Inner decoherence mechanisms are still present and not well identified.

Decoherence of **single spins in QDs** is better identified: the nuclear spin-bath.
Use of dots without nuclear spins, but S-T transitions should still be made possible.

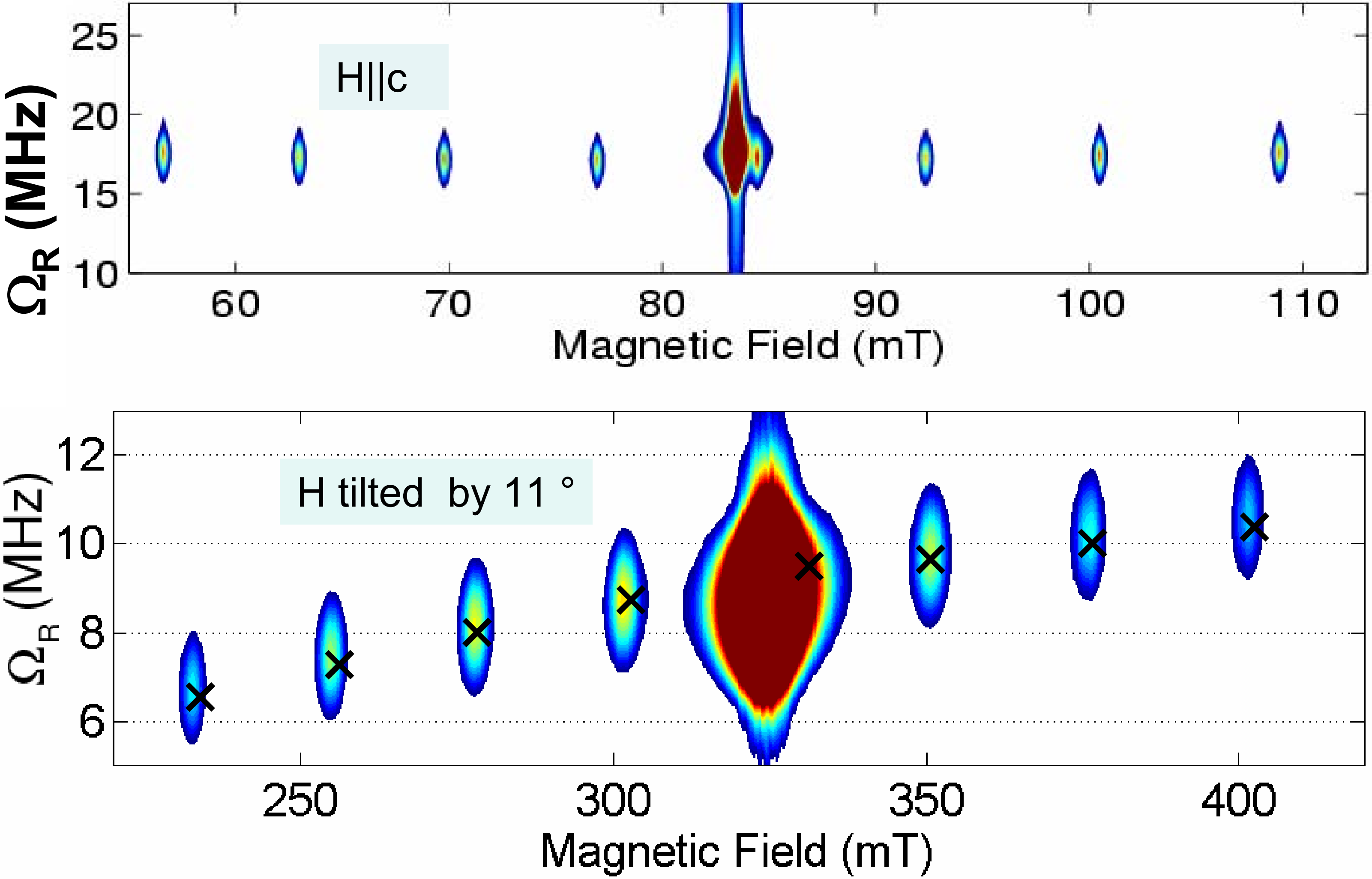
As well as with ensembles of spins, the **decoherence of ensembles of SQs or QDs qubit** (not yet studied, one exp.), should be much more drastic and should increase with the number of qubits (spin-bath and driven spin-bath, flux-bath and charge-bath).

The study of decoherence of ensembles of spins enables in particular the study of **driven decoherence.**

When interactions between ensembles of qubits become comparable to local quantum splittings, the **nuclear spin-bath** broadens the transition to the ordered state **suppressing quantum criticality** and limiting the lengthscale of entanglement.

Rabi oscillations of the 8 +1 electro-nuclear transitions

Er (0.001%):$CaWO_4$



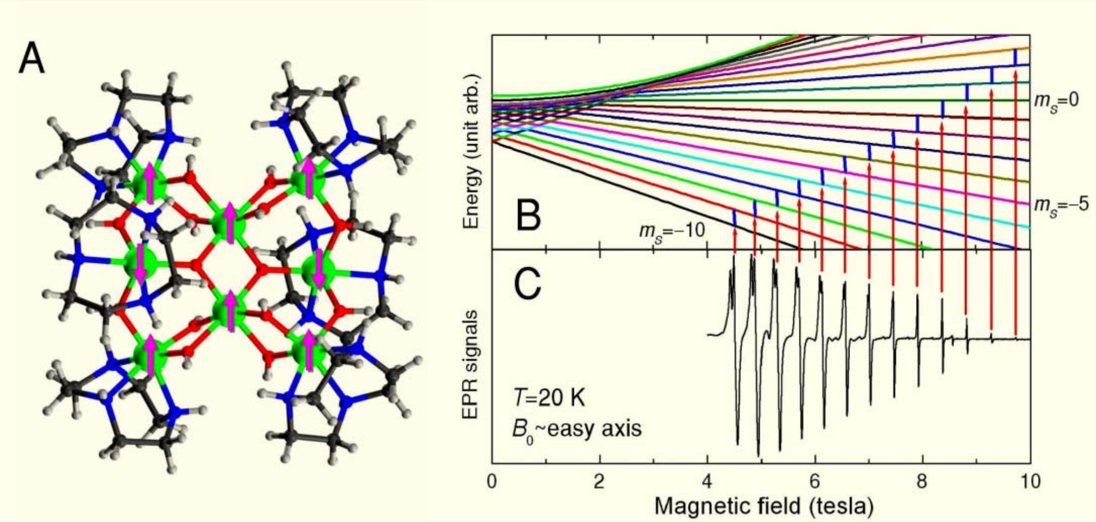
Large anisotropy of Rabi frequencies

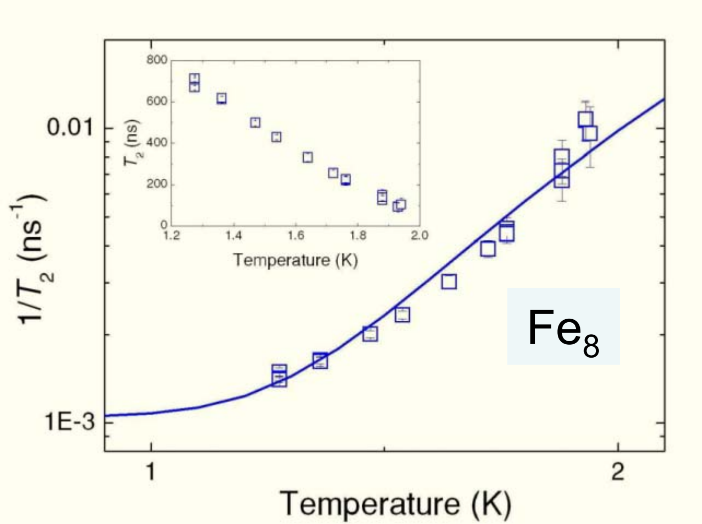
Phys. Rev. Lett. 24 Nov (2009)

More recent results with molecular magnets

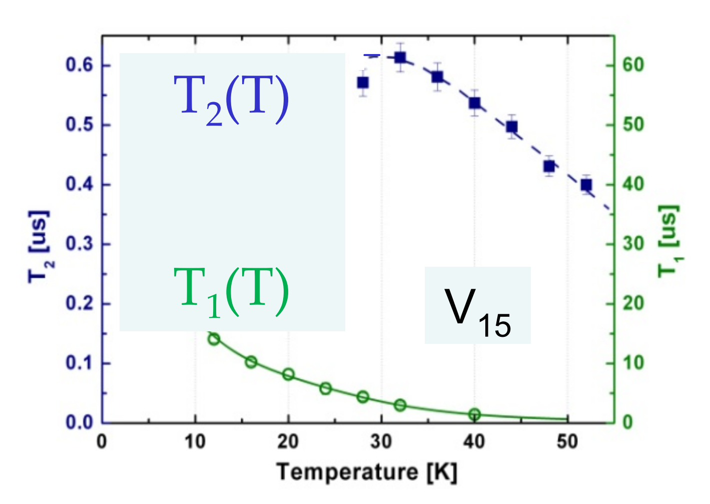
S. Takahashi et al, Santa Barbara and Tallahassee, Phys. Rev. Lett. (2009)

Fe_8: 8 spins 5/2 , Hilbert space dimension $D = 6^8 \sim 10^6$



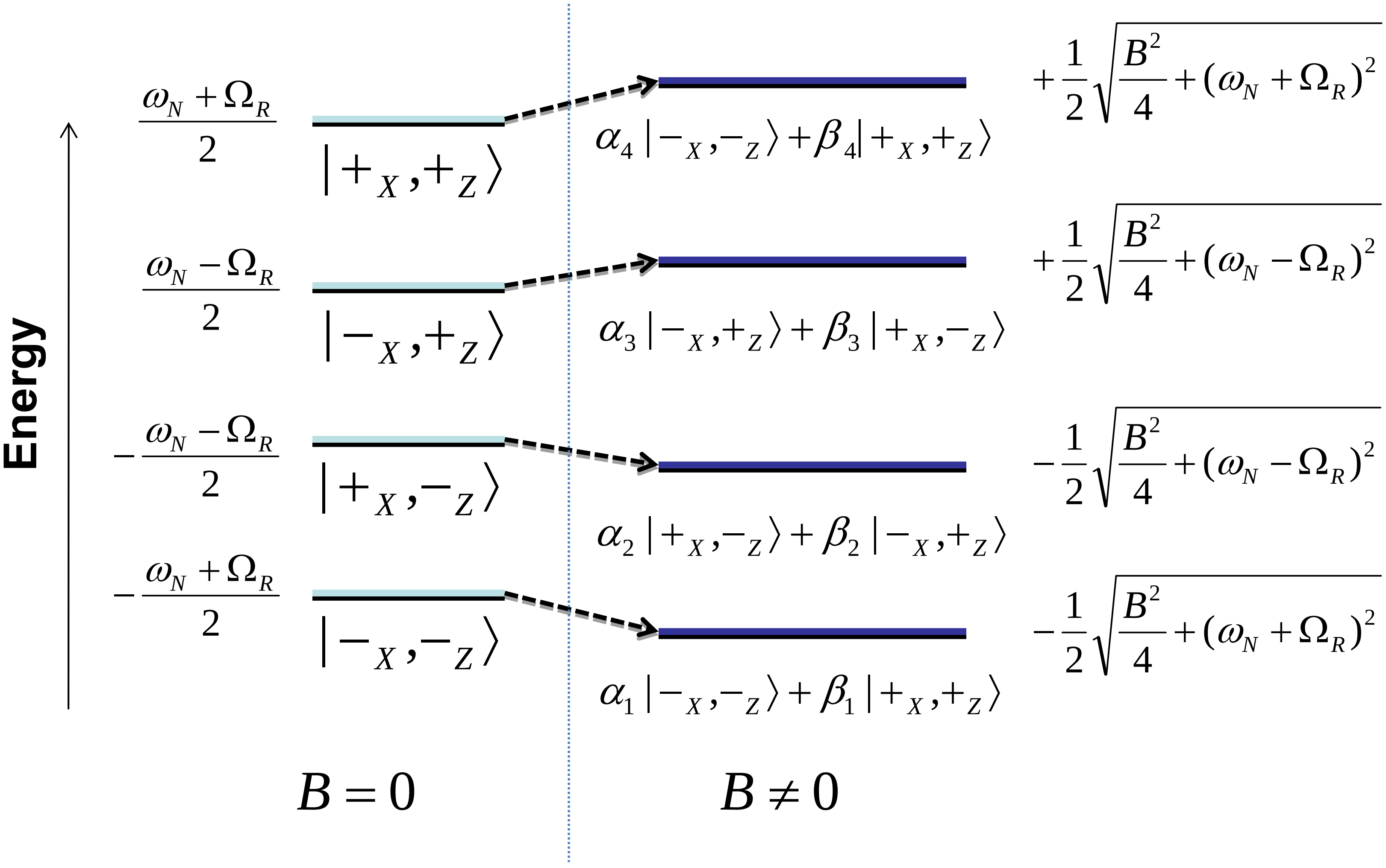


$$\frac{\hbar}{T_2} = C^{te} + \frac{\sqrt{\overline{E_{Dip}^2}}}{ch\left(\frac{\Delta_0}{kT}\right)} + \frac{\sqrt{\overline{E_{Ph}}}}{th\left(\frac{\Delta_0}{kT}\right)}$$



$$\mathrm{H}_{\mathrm{R}} = \omega_N I_Z + \Omega_R S_X + B S_Z I_X$$

$$(\omega_N \geq \Omega_R >> B > 0)$$



$\frac{\omega_N + \Omega_R}{2}$ $|+_X, +_Z\rangle$ → $\alpha_4 |-_X, -_Z\rangle + \beta_4 |+_X, +_Z\rangle$ $+\frac{1}{2}\sqrt{\frac{B^2}{4} + (\omega_N + \Omega_R)^2}$

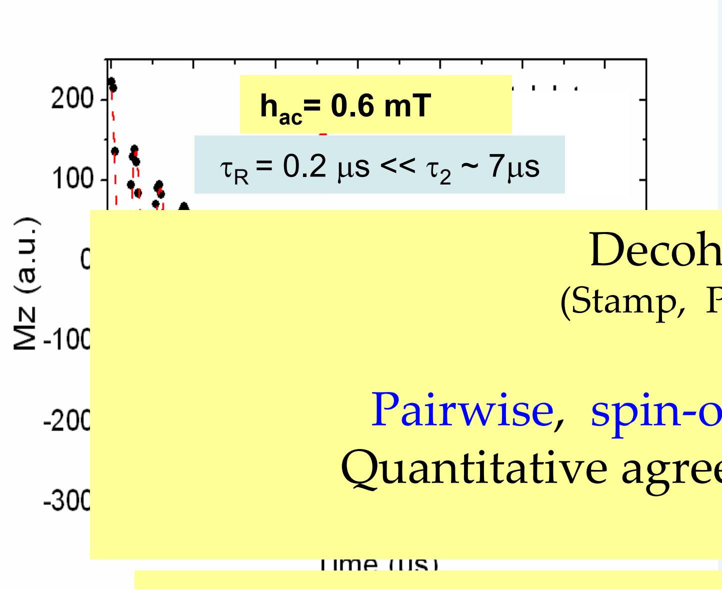
$\frac{\omega_N - \Omega_R}{2}$ $|-_X, +_Z\rangle$ → $\alpha_3 |-_X, +_Z\rangle + \beta_3 |+_X, -_Z\rangle$ $+\frac{1}{2}\sqrt{\frac{B^2}{4} + (\omega_N - \Omega_R)^2}$

$-\frac{\omega_N - \Omega_R}{2}$ $|+_X, -_Z\rangle$ → $\alpha_2 |+_X, -_Z\rangle + \beta_2 |-_X, +_Z\rangle$ $-\frac{1}{2}\sqrt{\frac{B^2}{4} + (\omega_N - \Omega_R)^2}$

$-\frac{\omega_N + \Omega_R}{2}$ $|-_X, -_Z\rangle$ → $\alpha_1 |-_X, -_Z\rangle + \beta_1 |+_X, +_Z\rangle$ $-\frac{1}{2}\sqrt{\frac{B^2}{4} + (\omega_N + \Omega_R)^2}$

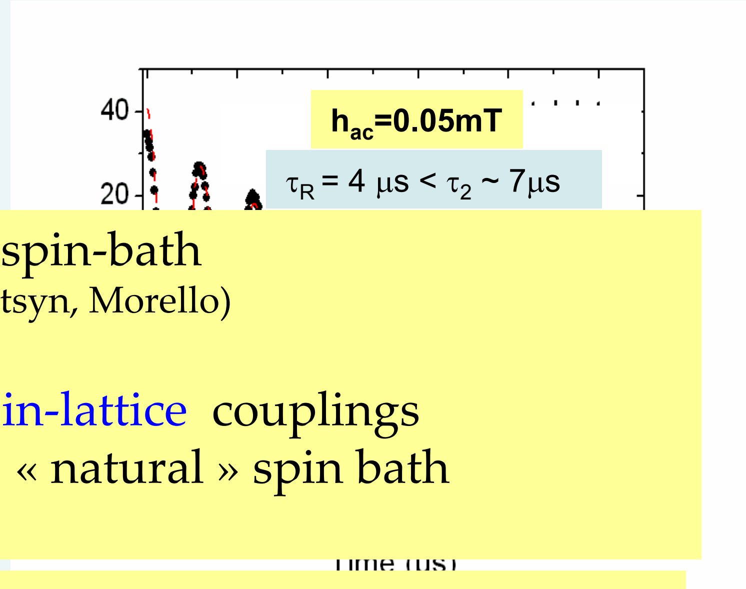
$B = 0$ $B \neq 0$

Damping of oscillations
Effect of the microwave power



h_{ac}= 0.6 mT

τ_R = 0.2 µs << τ_2 ~ 7µs



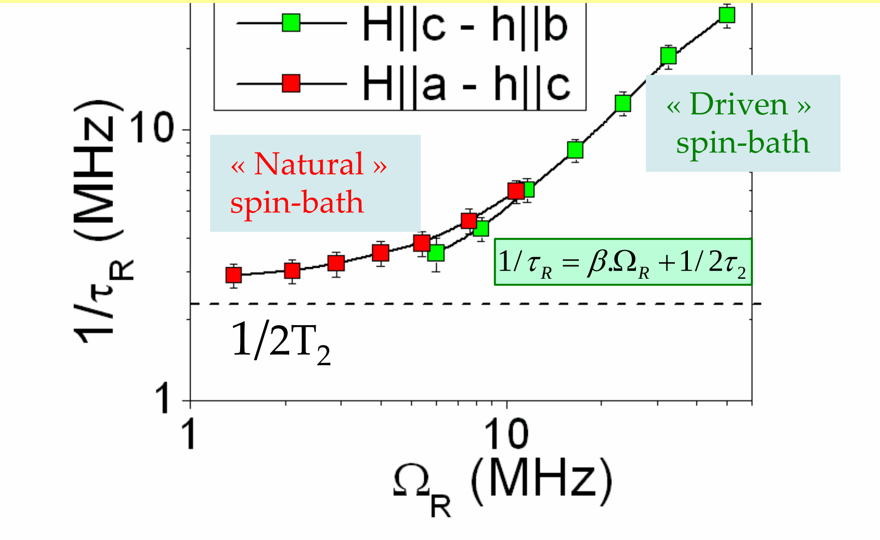
h_{ac}=0.05mT

τ_R = 4 µs < τ_2 ~ 7µs

Decoherence: the spin-bath
(Stamp, Prokof'ev, Tupitsyn, Morello)

Pairwise, spin-orbit and spin-lattice couplings
Quantitative agreement with « natural » spin bath

Study of the spin-bath + microwaves is needed
(also for superconducting qubits, when N > 1)



« Natural » spin-bath

« Driven » spin-bath

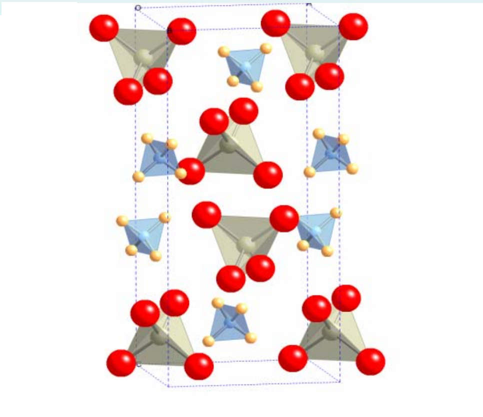
$1/\tau_R = \beta.\Omega_R + 1/2\tau_2$

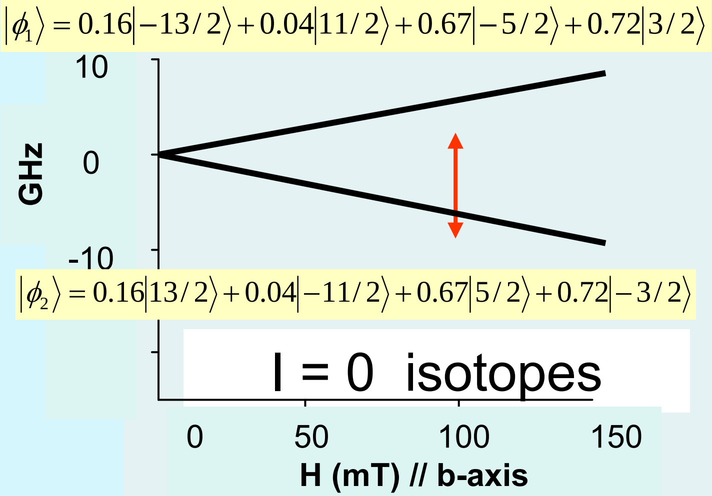
$1/2T_2$

Rare-earth ions: **Er^{3+}:$CaWO_4$**

Two isotopes: I=0, I=7/2

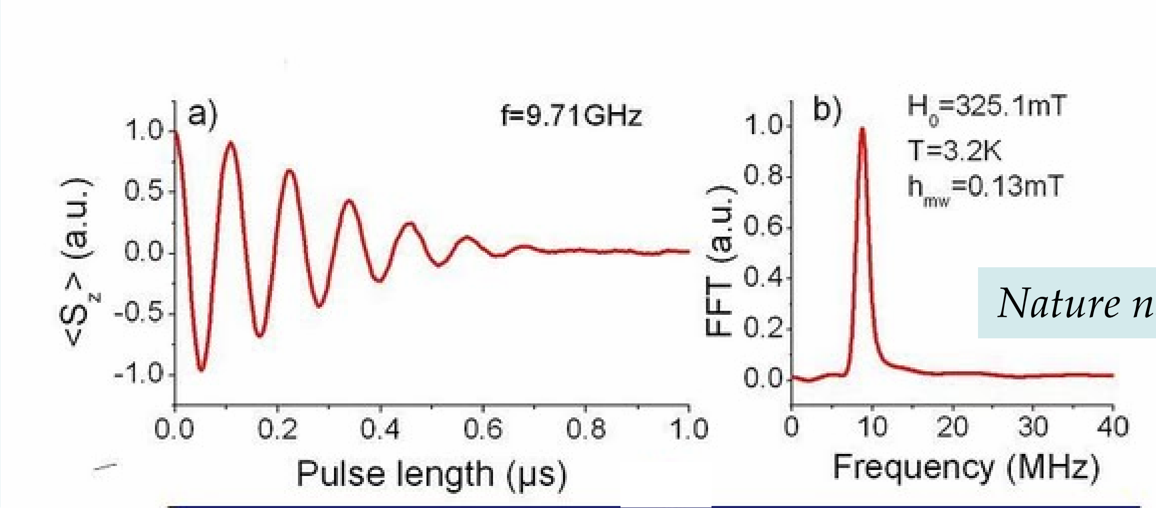
$$H_{CF} = \alpha_J B_2^0 O_2^0 + \beta_J (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma_J (B_6^0 O_6^0 + B_6^4 O_6^4)$$



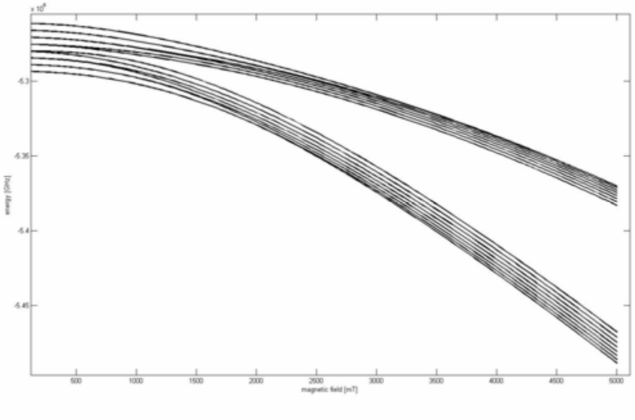


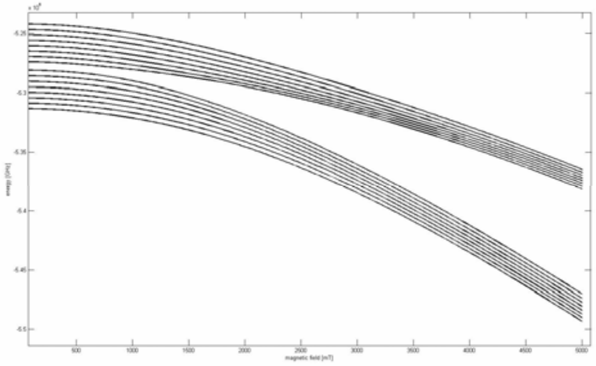
$$|\phi_1\rangle = 0.16|-13/2\rangle + 0.04|11/2\rangle + 0.67|-5/2\rangle + 0.72|3/2\rangle$$

$$|\phi_2\rangle = 0.16|13/2\rangle + 0.04|-11/2\rangle + 0.67|5/2\rangle + 0.72|-3/2\rangle$$



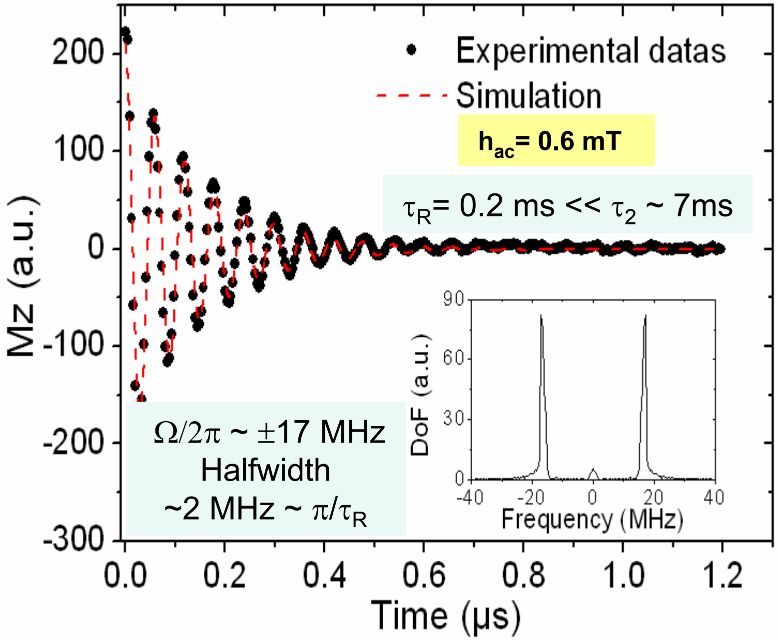
Nature nanotechnology, 1 Jan (2007)


x 10^{4}
-5.3
-5.35
-5.4
-5.45
energy [GHz]
500
1000
1500
2000
2500
3000
3500
4000
4500
5000
magnetic field [mT]


x 10^{4}
-5.25
-5.3
-5.35
-5.4
-5.45
-5.5
energy [GHz]
500
1000
1500
2000
2500
3000
3500
4000
4500
5000
magnetic field [mT]

Damping of oscillations

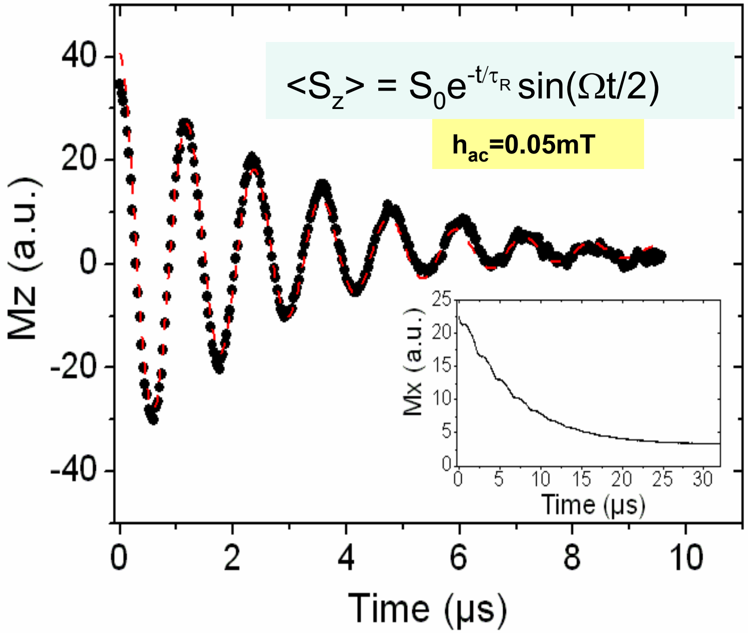
Effect of the microwave power (0.05% $Er:CaWO_4$)



h_{ac}= 0.6 mT

τ_R= 0.2 ms << τ_2 ~ 7ms

$\Omega/2\pi$ ~ ±17 MHz
Halfwidth
~2 MHz ~ π/τ_R



$\langle S_z \rangle = S_0 e^{-t/\tau_R} \sin(\Omega t/2)$

h_{ac}=0.05mT

The damping rate decreases with microwave power

Stochastic noise, Interferences

No-interactions, reversible decoherence

Nature nanotechnology (2007)

Tu me dis que le SQUID est arrêté pendant la manipulation des qubits, mais tu le mets en m

Et Ib commence à augmenter après la variation adiabatique de flux ?

gmentation du Ib (cad la rampe montee du pulse de lecture) genere une variation NON-adia
gique, dans un point "non-magique" ou un peu faire tranquille leur lecture.

Mais de toutes les façons les fils sont tous supra et il y s des plasmons dans les fils, n'est-ce

a le bruit electronique Johnson-Nyquist. Le circuit contient des resistances au dela des fils s
erreur de mesure car the Ic qu'on va mesurr ne sera pas le réel (fidelite).
s ce bruit (aussi lié au piégeage de charges dans la barriere (ce qui change la résistance d
plus : le bruit en question est d'origine thermique il est donc plutôt dans les fils haute Temp
SQUIDs donnent du bruit en 1/f (mais en g fequences < 1Hz)

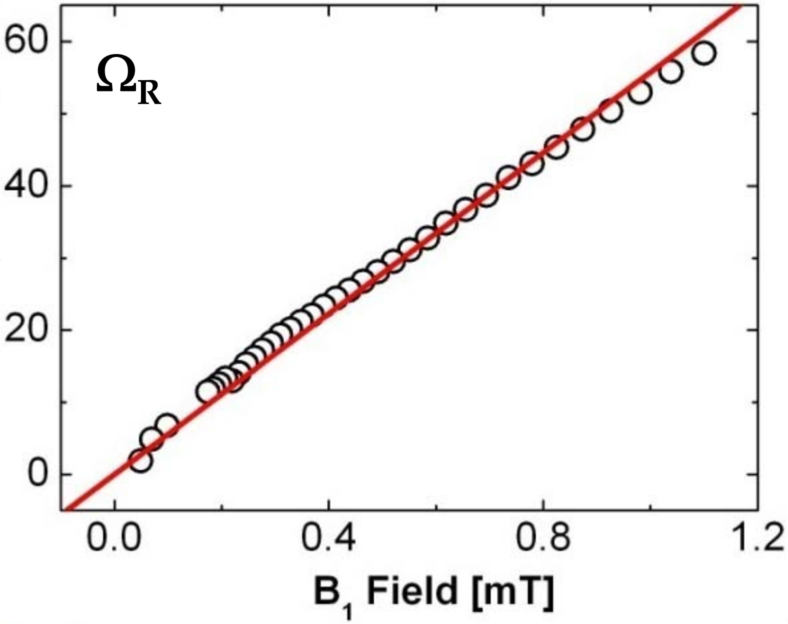
erait sans limites.

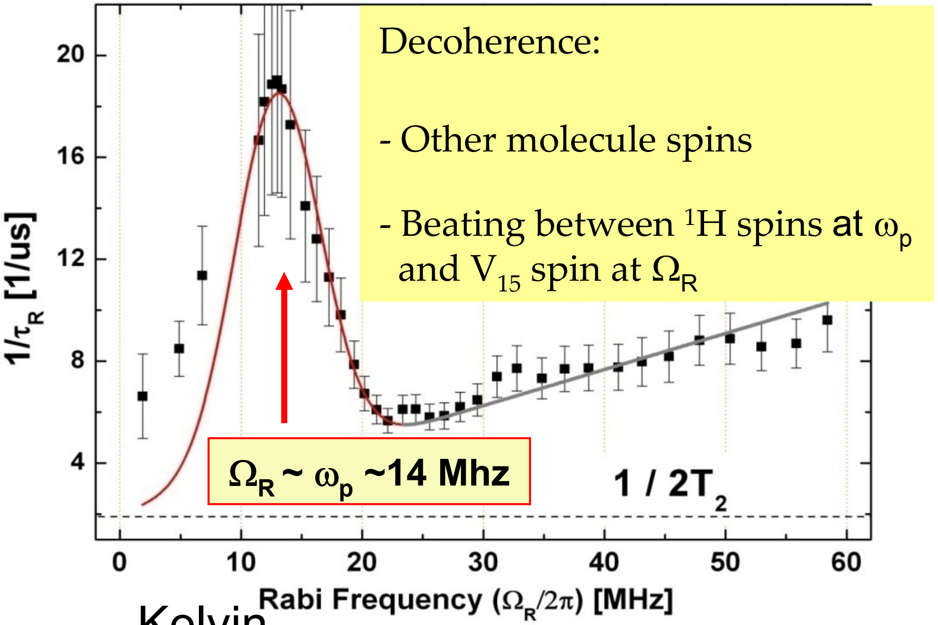
e first experimental coherent manipulation of flux qubits .."
postdoc sur le plan de construire le qubit et la demonstration du gap tunnel (incoherent).

J'ai pas compris l'explication avec la phase du squid. Moi, je le vois plus simple. Le squid e
re du qubit. Le SQUID detecte si ce moment magnetique geant est "up" ou "down". Comme
le up ou down), le cosine varie.Je suis tout à fait d'accord avec ça!
je ne sais plus ou.
tat antisymmetrique /up> - /down> est différent tandis que celui del'état symmétrique /up>
0.

t le premier nul. Mais c'estr bien vrai que dans les dux cas le moment est de signe opposé

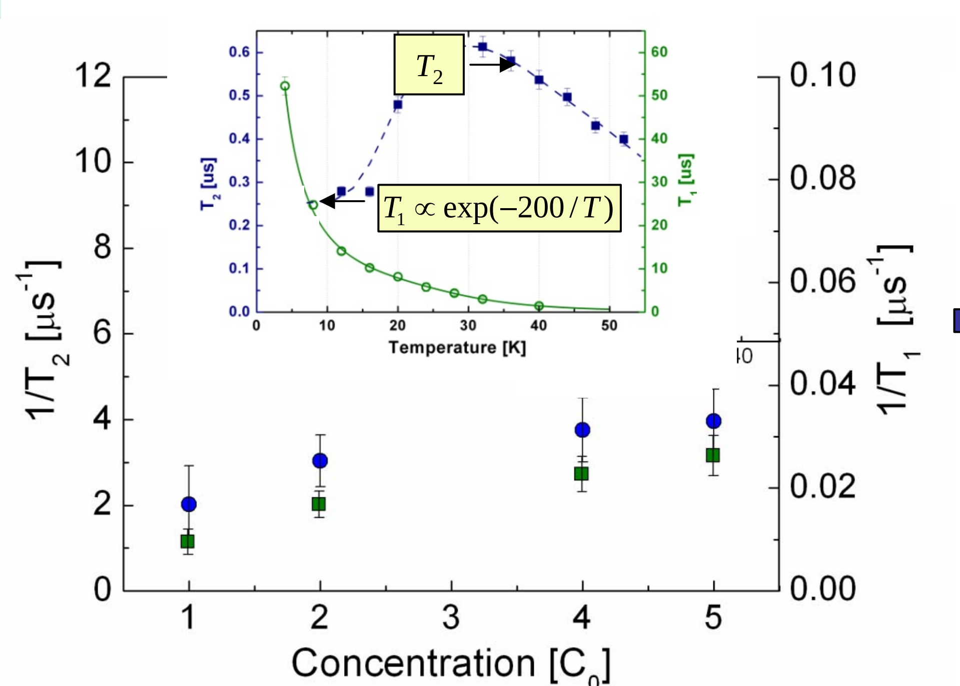
Rabi oscillations of a SMM vs mw-field and temperature

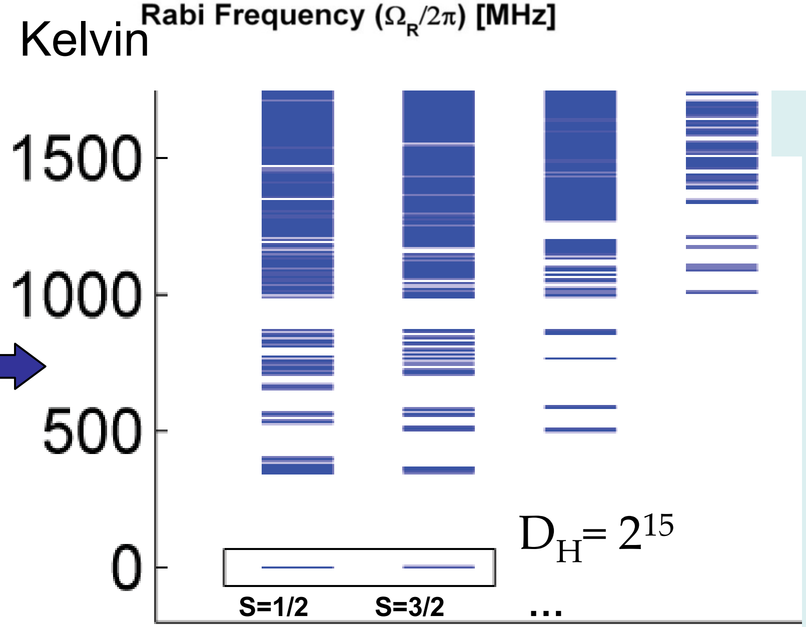




Decoherence:

- Other molecule spins
- Beating between ^1H spins at ω_p and V_{15} spin at Ω_R

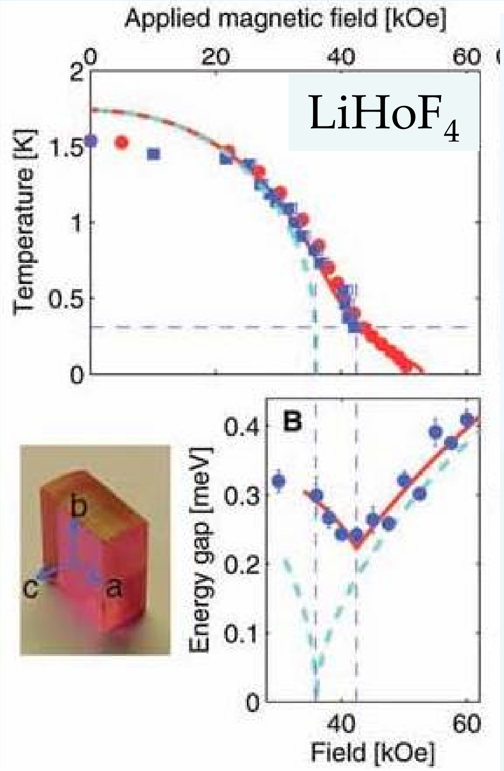


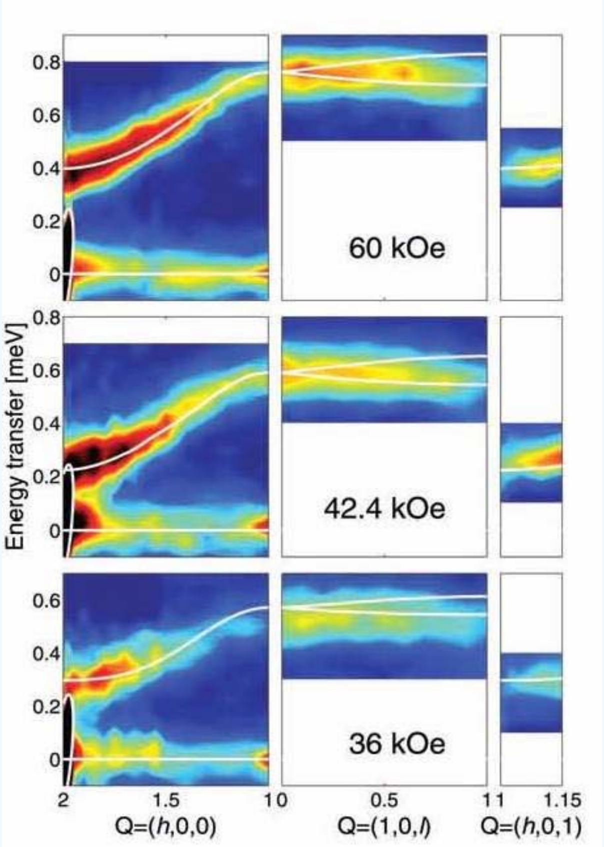


$D_H = 2^{15}$

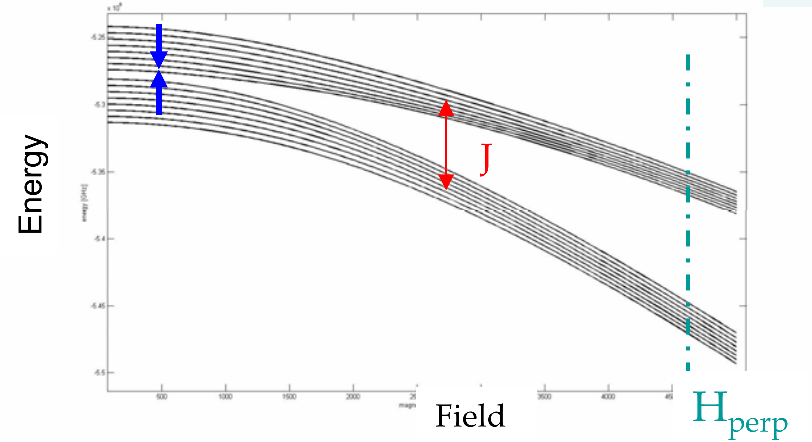
Quantum phase transition of a magnet in a spin-bath

H. Ronnov, R. Parthasarathy, J. Jansen, G. Aeppli, T. Rosenbaum, D. McMorrow, Science, 15 April 2003





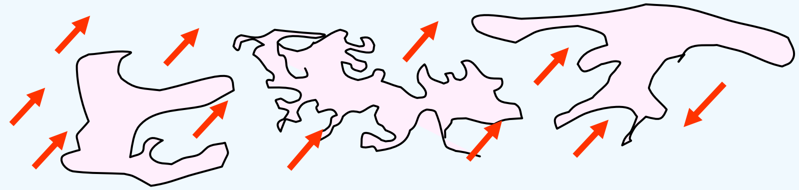
Single flips !



$\Delta_2 > \Delta_1 >$ J ⟶ Q-Fluct. (Δ_1, Δ_2) Q-« Para »

$\Delta_2 >$ J $> \Delta_1$ ⟶ Q-Fluct. (Δ_2) Q-Dynamics ↘, clusters

J $> \Delta_2 > \Delta_1$ ⟶ Q-Fluct. (Nuclear only), ordered state.

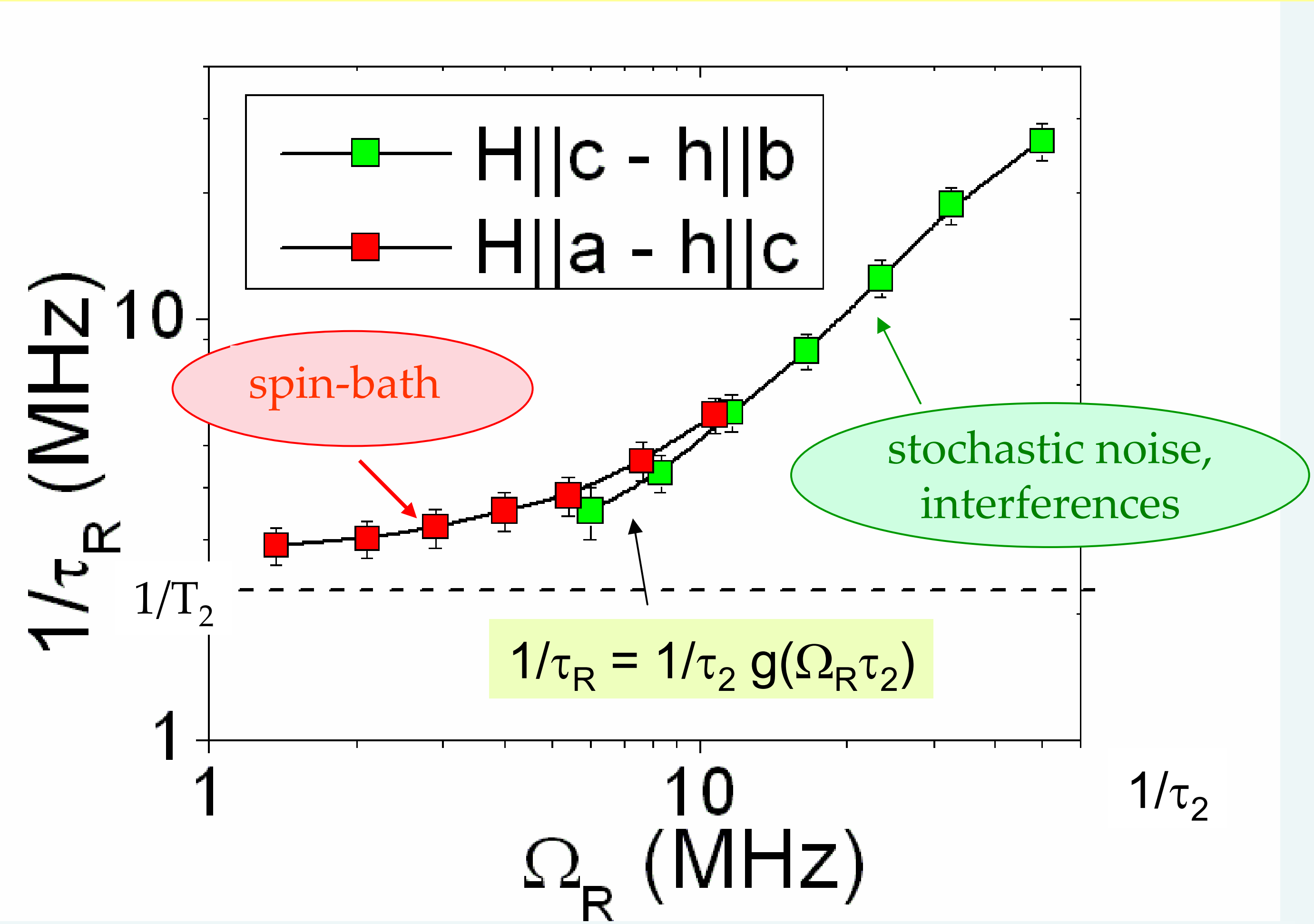


Q. Phase transitions:
Decoherence (by e.g. kT) kills the superconding PT

In SQUID reade-out : also a QPT: le SQUID switches from super to normal,
But the QPT is not induced by decoherence beacuse it is the probe which
detects the states of coherence. QPT is a part of the measurement.

Spins systems : OK to test the effect of decoherence in QPT:
LiHoF4: Aeplli dit que c’est le bain de spin qui empêche la divergence ferro du système.
Maisz le coiplage hyperfin est fort et les pins nucleau ires font pat rtie dud d système:
donner un cycle d’hystéresis du système dilué et dire que ça ne cham ge pas si conce,ntr

The damping rate scales with the Rabi frequency



spin-bath

stochastic noise, interferences

$1/\tau_R = 1/\tau_2 \, g(\Omega_R \tau_2)$

Assuming that each spin experiences a stochastic field oscillating at the frequency ω (Shakhmuratov et al, Phys. Rev. Lett., 1997)

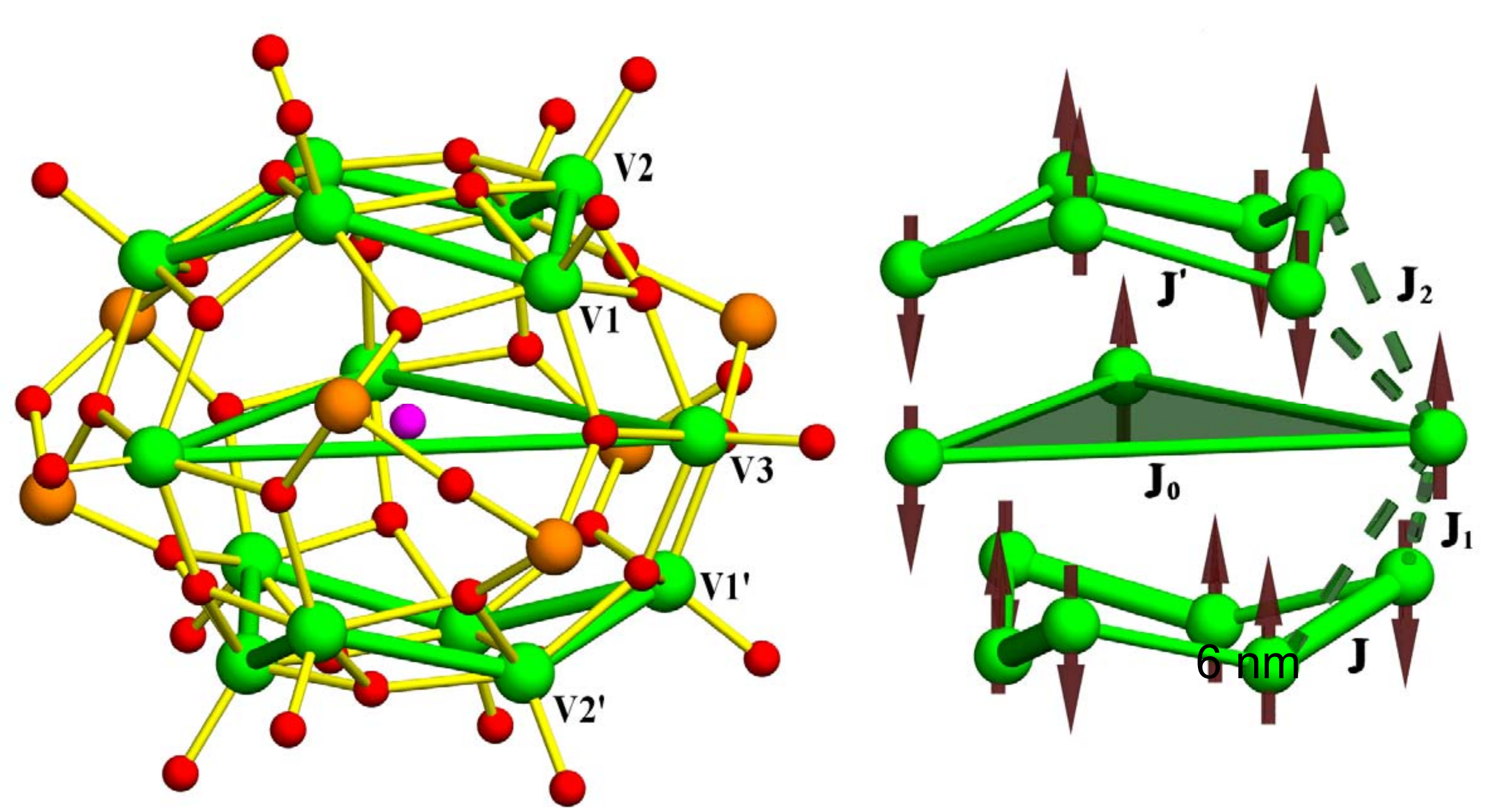
$$1/\tau_R = \beta.\Omega_R + 1/2\tau_2$$

Nature nanotechnology (2007)

Rabi oscillations of a molecular magnet

$$V_{15} \equiv \left[V_{15}^{IV} As_6^{III} O_{42} \left(H_2O\right)\right]^{6-}$$

S=1/2

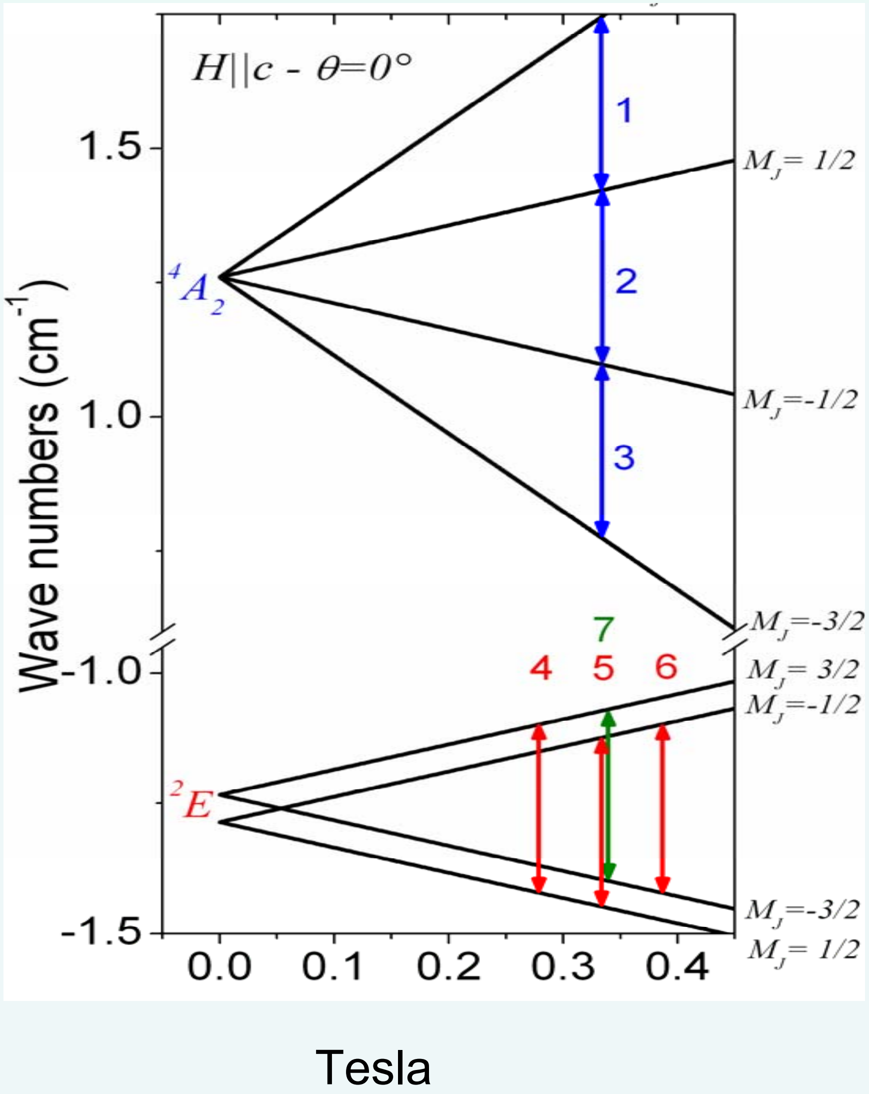


These molecules are wrap up with a surfactant and dispersed in a Chloroform solution

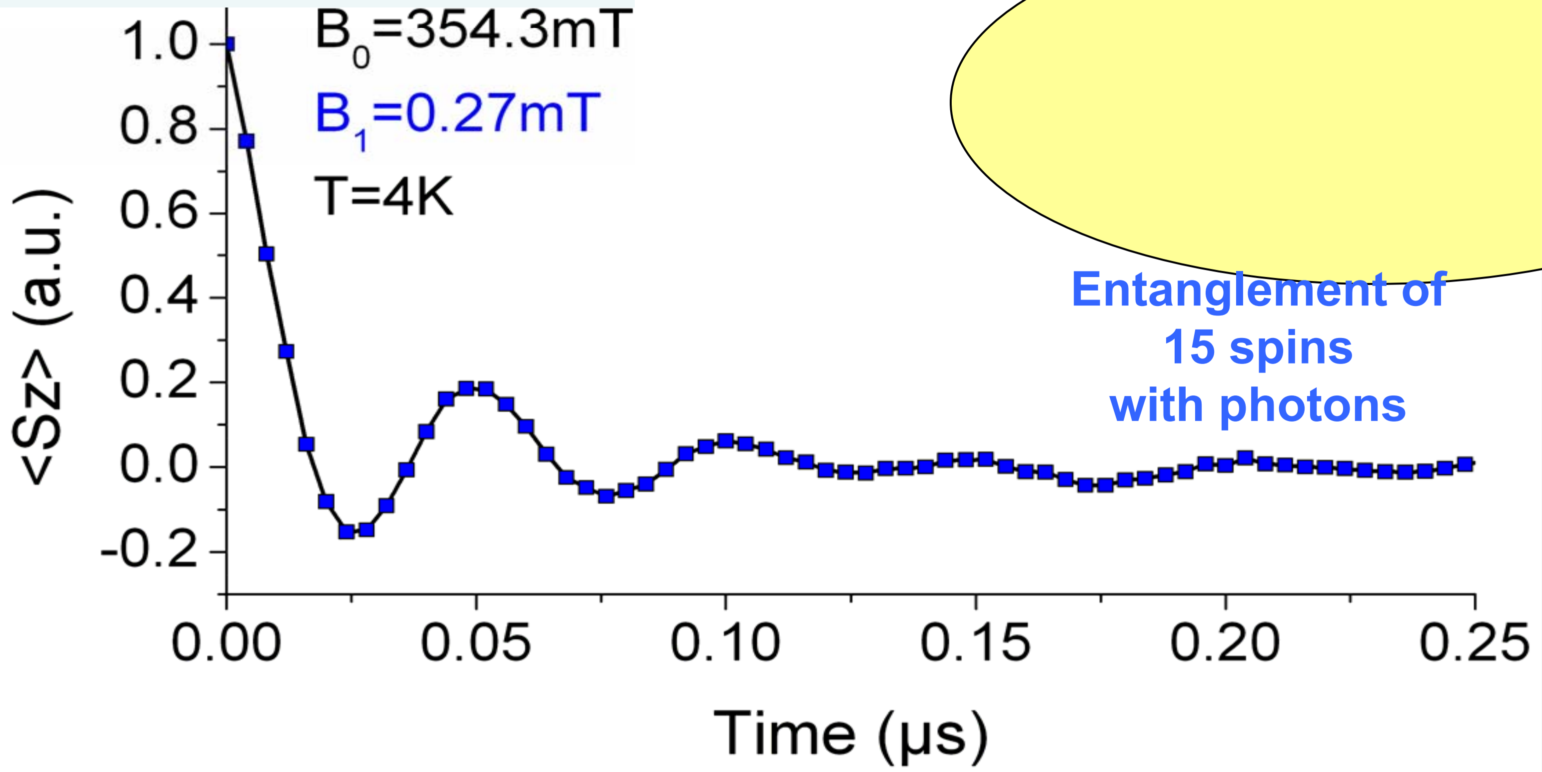
Collaboration Achim Müller, Univ. Bielefeld

Complex Hamiltoninan

$$H = -J_0 \sum_{\substack{i,j=1 \\ (i<j)}}^{3} \boldsymbol{S}_i \boldsymbol{S}_j + \sum_{ij=12,13,31} \boldsymbol{D}_{ij} \left(\boldsymbol{S}_i \times \boldsymbol{S}_j \right) + A \sum_{i=1}^{3} \boldsymbol{I}_i \boldsymbol{S}_j + g\mu_B \boldsymbol{H} \sum_{i=1}^{3} \boldsymbol{S}_i$$



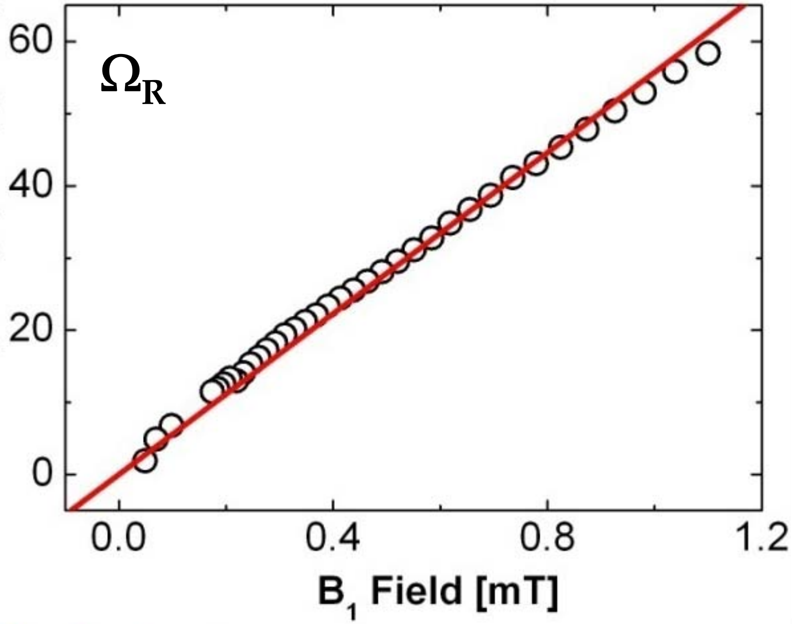
First Observation of Rabi Oscillations in a Molecular Magnet (V_{15})

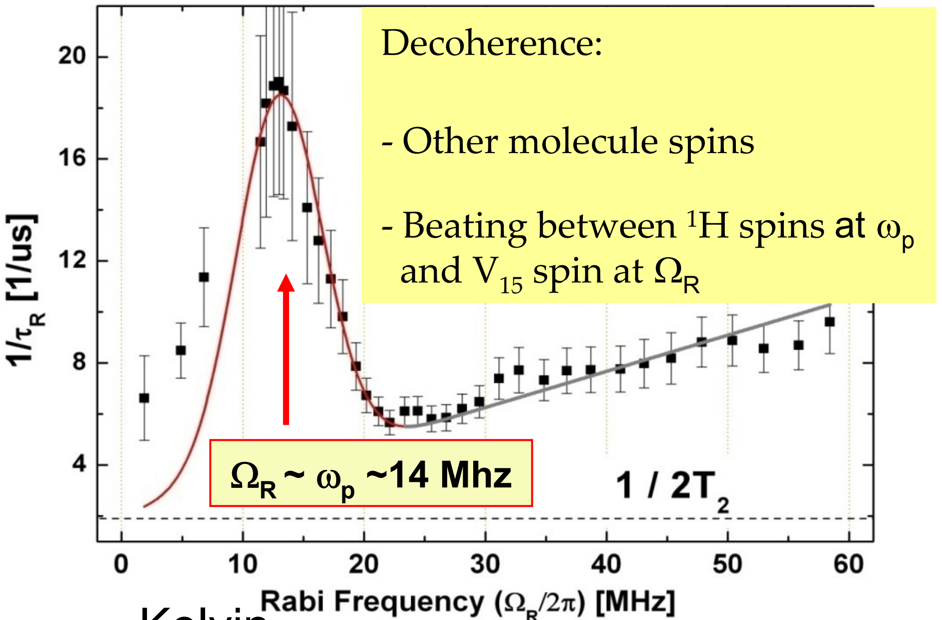


Entanglement of 15 spins with photons

Nature, 8 May (2008).
(see also : P. Stamp, *News* & *Views* , same issue).

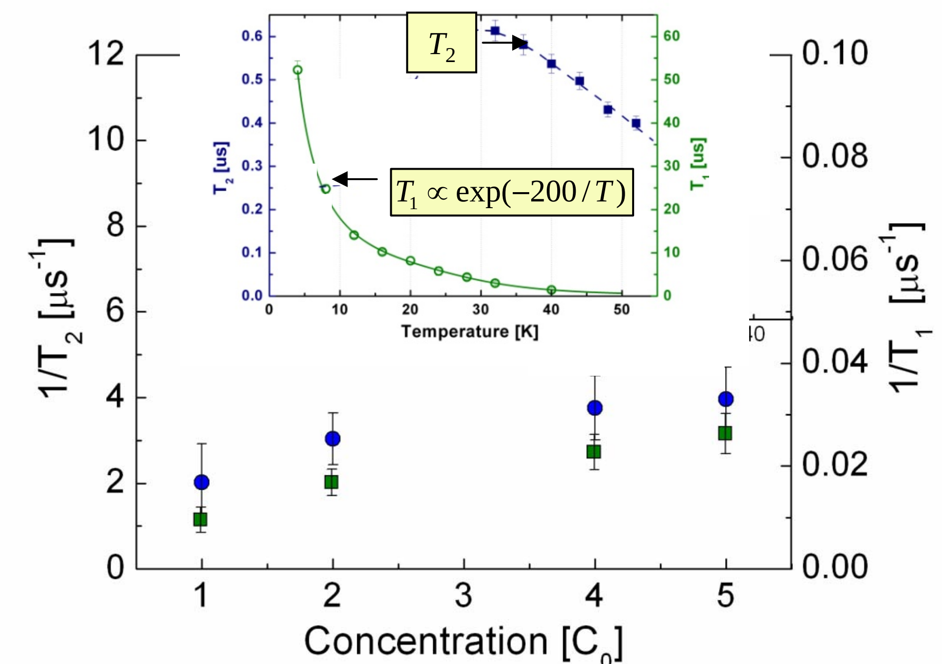
Rabi oscillations of a SMM vs mw-field and temperature

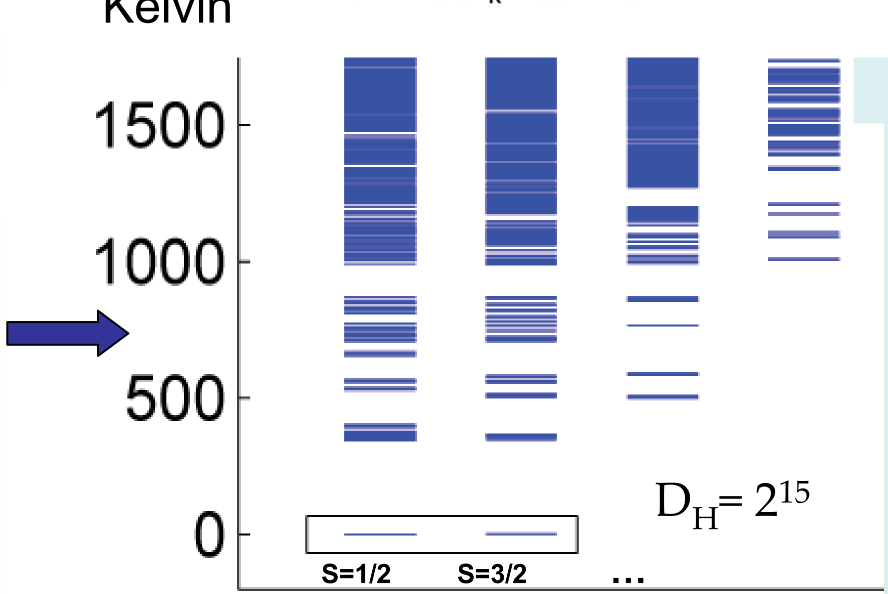




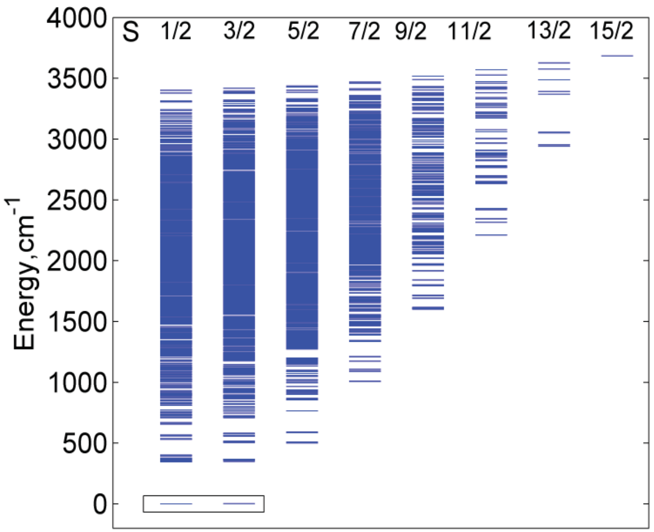
Decoherence:

- Other molecule spins
- Beating between ^{1}H spins at ω_p and V_{15} spin at Ω_R





$D_H = 2^{15}$

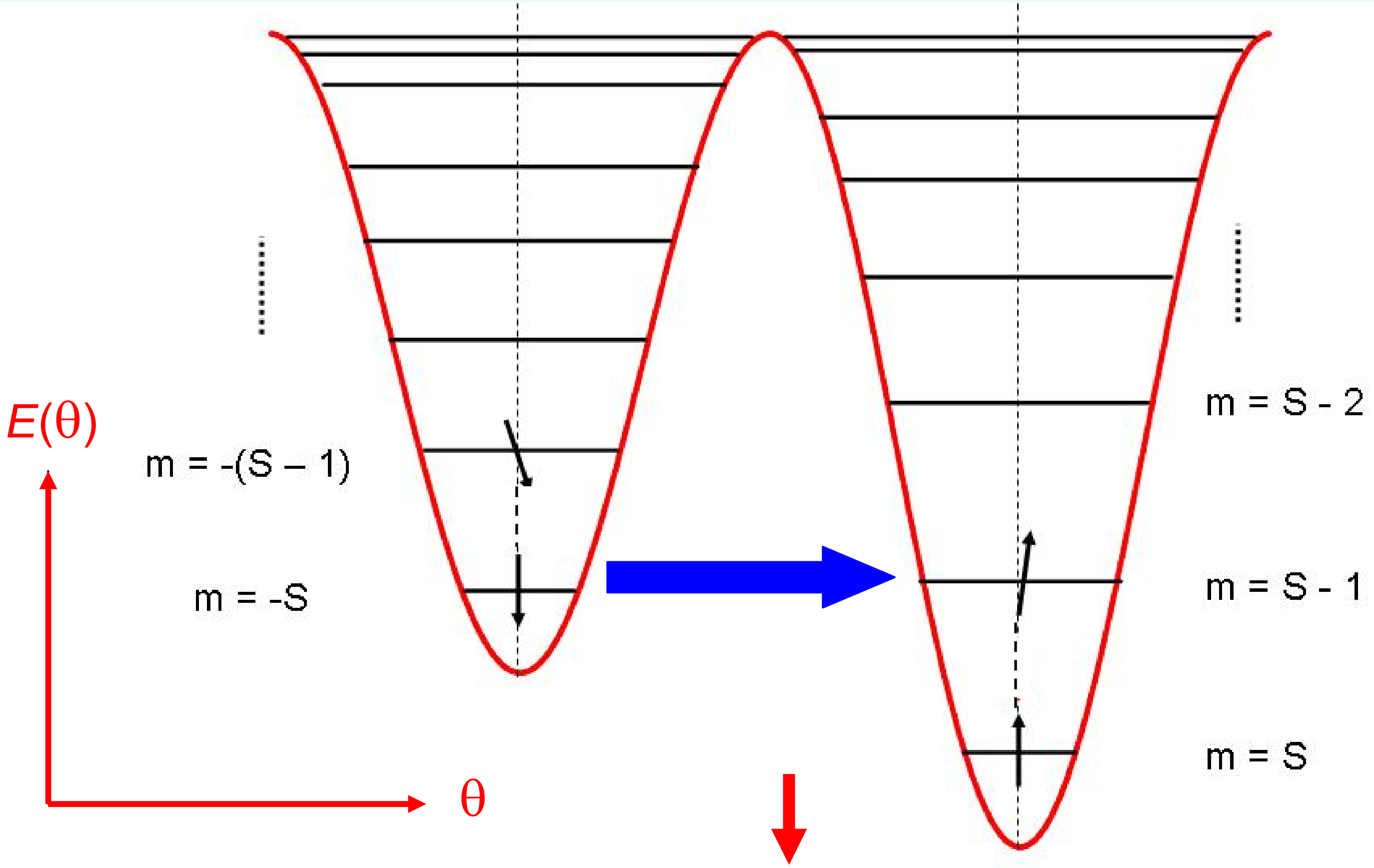


Energy, cm^{-1}
4000
3500
3000
2500
2000
1500
1000
500
0
S
1/2
3/2
5/2
7/2
9/2
11/2
13/2
15/2

Quantum tunneling of SMMs (Mn12-ac type)

$$H = -DS_z^2 + BS_z^4 \ldots - g\mu_B S_z H_z + B(S_x^2 + S_y^2)\ldots$$

$E(\theta)$ with $\cos\theta = \langle S_z \rangle / S$, $\langle S_z \rangle = m$

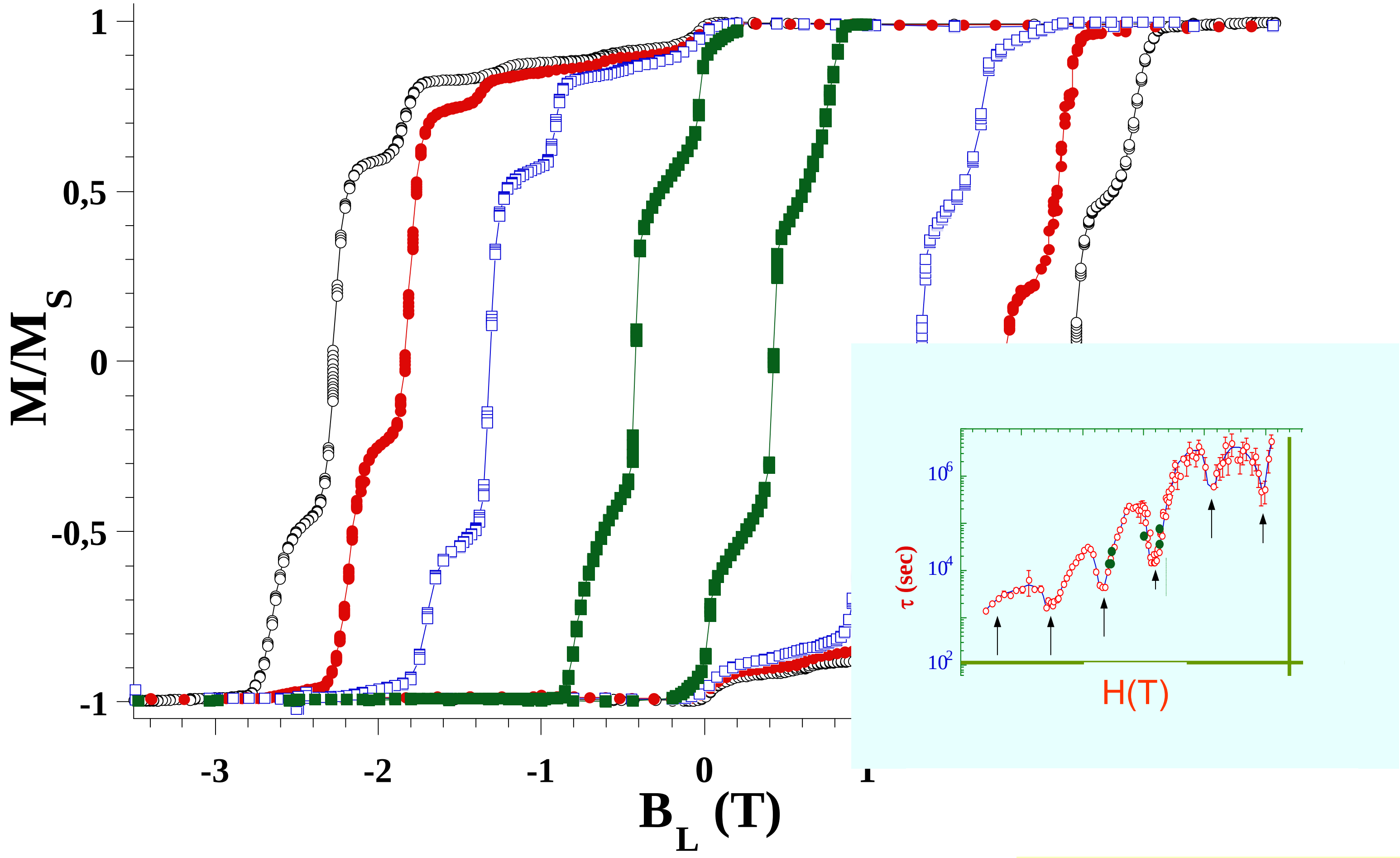


Magnetization steps and quantum relaxation

Resonant tunneling of magnetization (Mn_{12}-ac, S=10)

Quantum tunneling and classical hysteresis

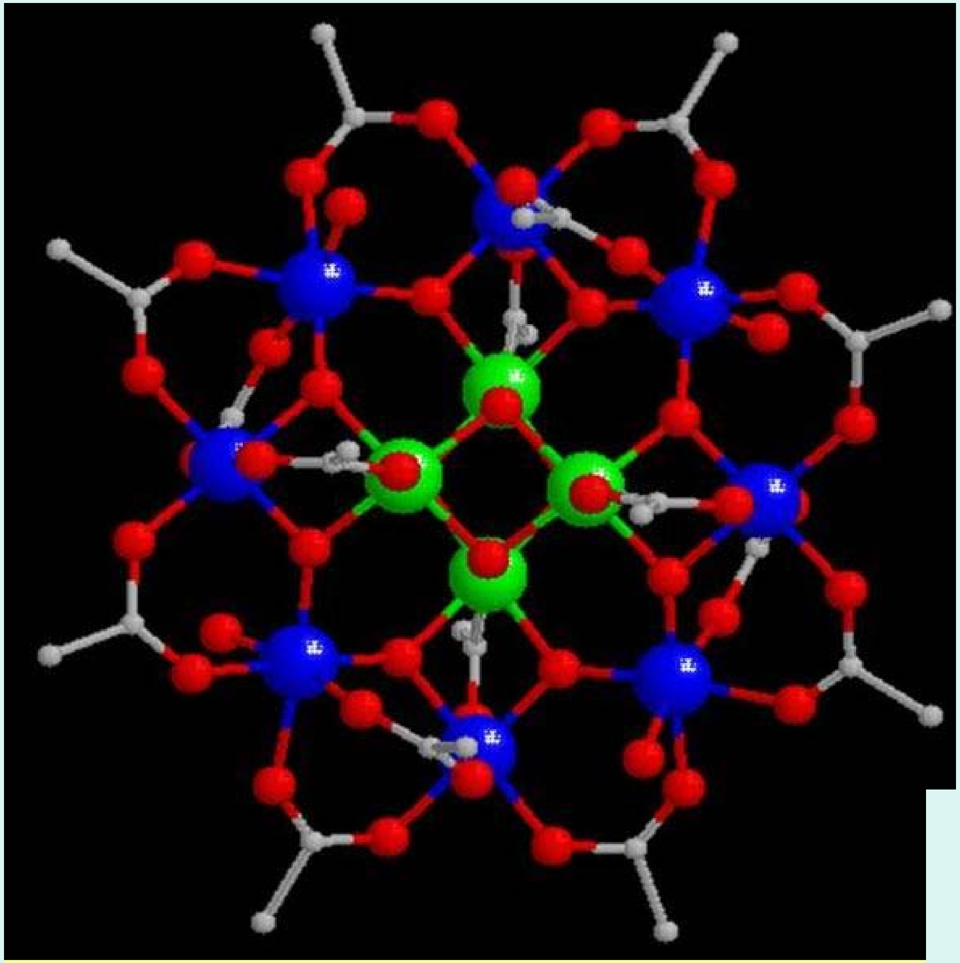
Hysteresis loop



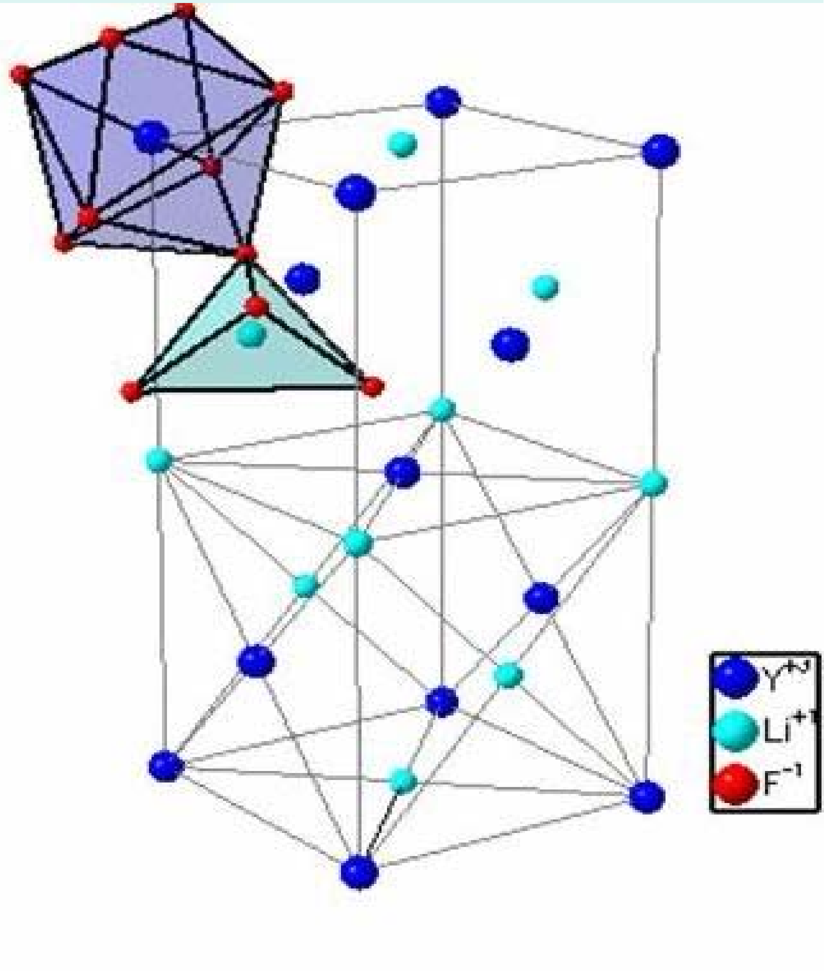
$H_n = nD/g\mu_B \sim nH_A/2S$

Nature, 12 Sept (1996)

Fom SMMs to simple paramagnetic ions (RE-ions)



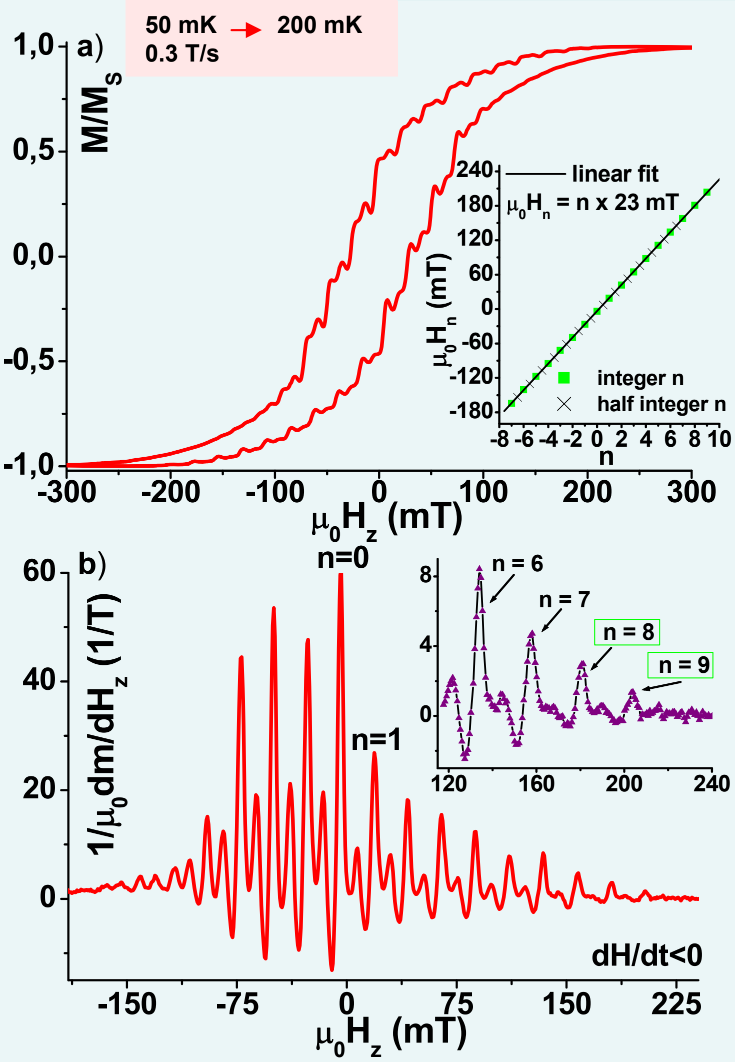
Nature 12 Sept (1996)

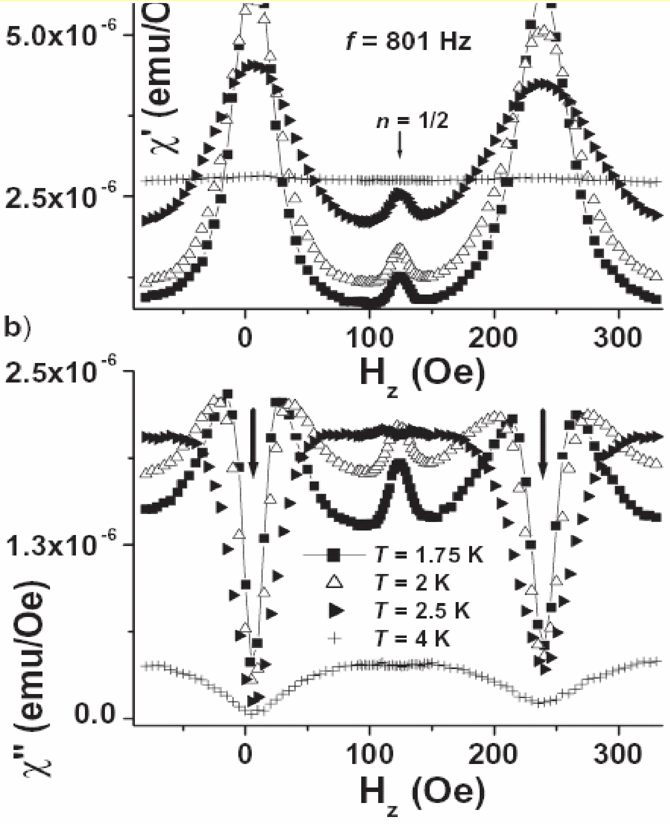


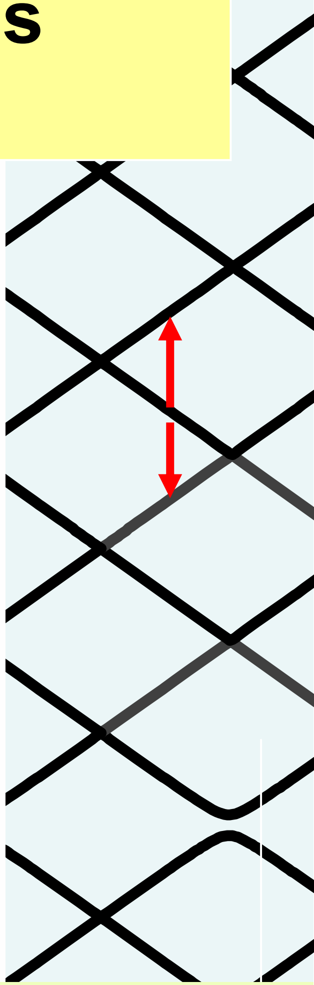
Phys. Rev. Lett. 17 July (2001)
Phys. Rev. Lett. 19 Dec (2003)

Additional steps at intermediate fields

Fast measurements







Simultaneous tunneling of Ho^{3+} pairs
(4-bodies tunnelling)

Detailed studies in ac-susceptibility.
Accurate fits of many-body tunnel relaxation with spins-spins, spin-phonons, bottleneck, weak CF disorder (B.Malkin). **PRB, 74, 184421 (200**

Coherent quantum regime

Pulsed EPR measurements

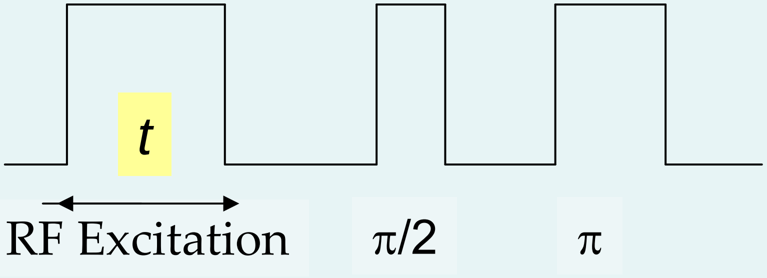
X band spectrometer (9-10GHz)

Continuous wave (CW)

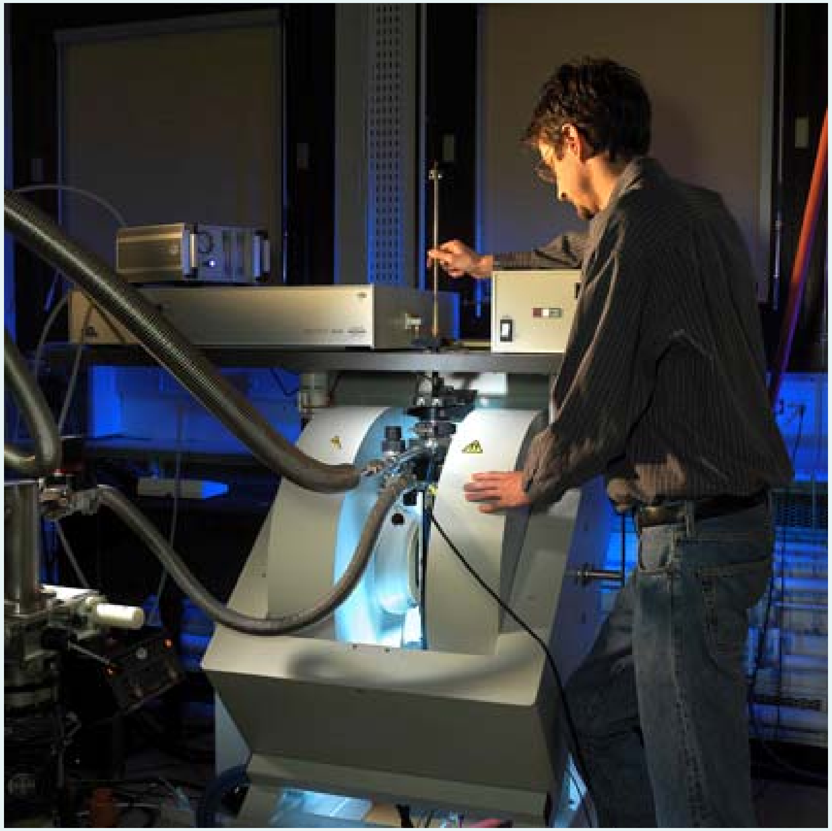
Time resolved (TR) or pulsed

Temperature 2.5K to 300K

EPR sequence used



$$\text{Echo} \propto \langle S_z \rangle = f(t)$$



Bruker Elexys E580

Copyright CEA-Grenoble

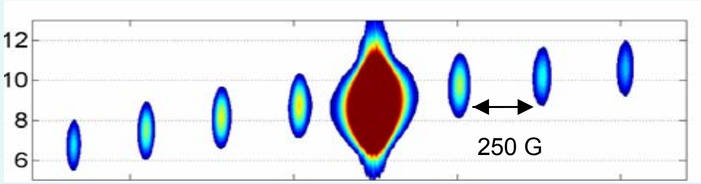
Rare-earths ions (Er)
Single Molecule Magnets (V_{15})

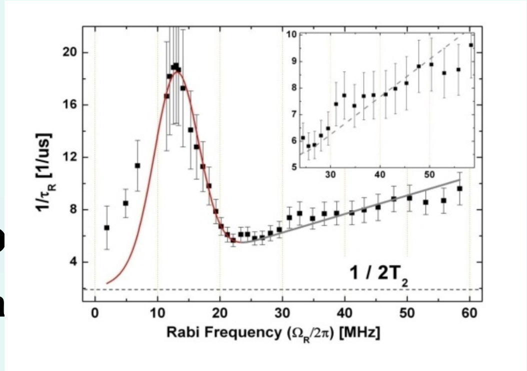
Conclusion

Demonstration of the existence of two new types of spin qubits

1- «**The spin-orbit qubits**» in rare-earth ions (and any other system with large spin-orbit coupling).

- The point symmetry of the matrices influences deeply the Rabi oscillations
- New ways to manipulate quantum oscillations in e.g. QCs
- Coherence times reaching the millisecond at 4K.





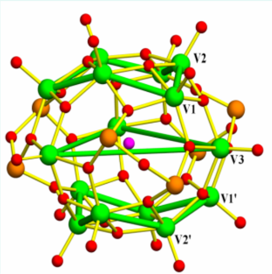
2- The «**Nanometer-size spin qubits** » in Single Mo

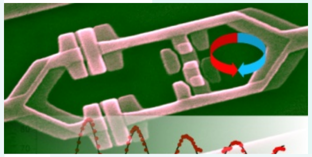
- Quantum oscillations are compatible with com spaces, complex interactions)
- Decoherence by nuclear spins. Especially by protons ($\Omega_R < 20$ MHz)
- Coherence times must be improved (should be possible)

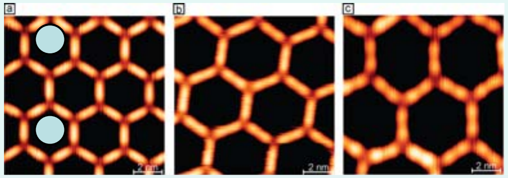
Next future:

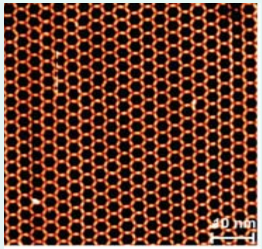
1- Detailed studies of decoherence in both types of systems

2- From 3D to 2D and 0D systems (films, single-objects)









From: M. Ruben, J. V. Barth et. al., INT Karlsruhe, TU Munich

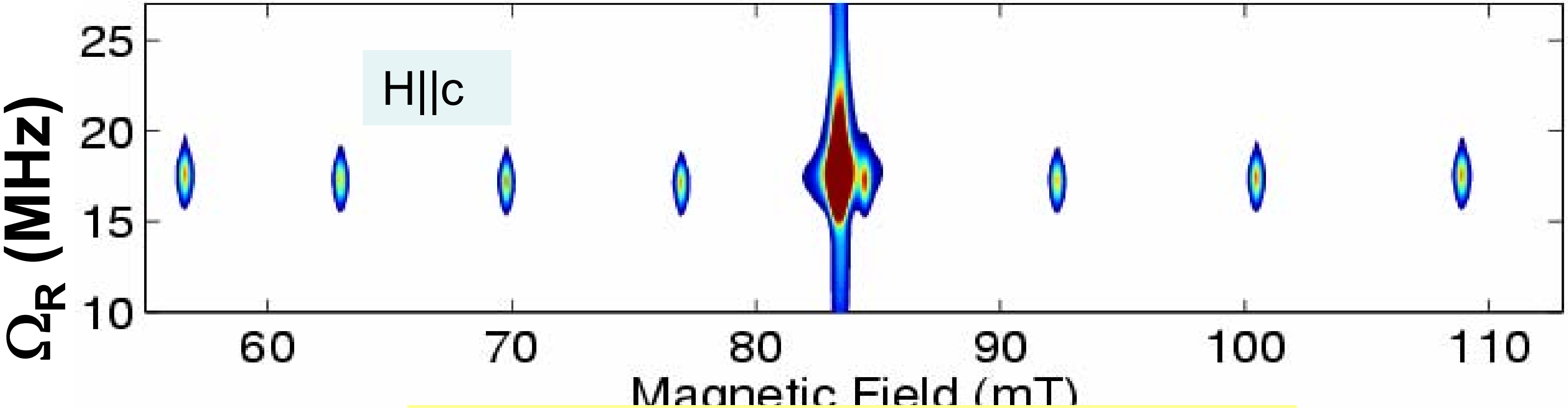
Collaborations

Quantum coherence of SREs and SMMs

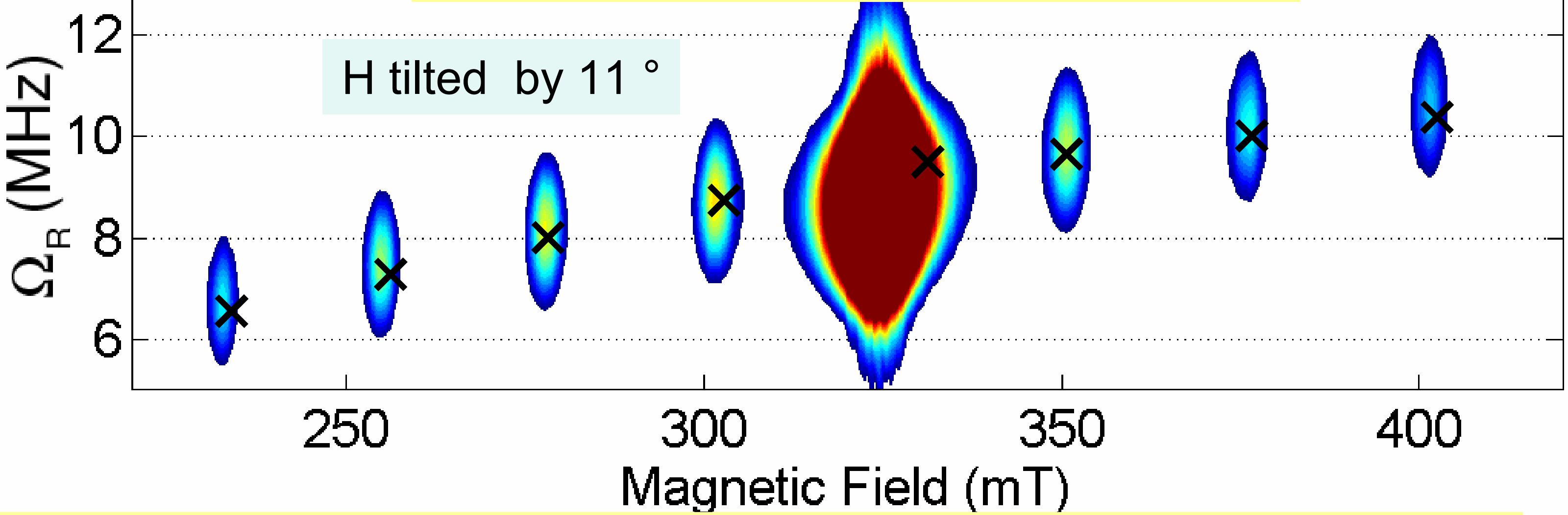
Physics: S. Gambarelli (CEA-Grenoble), J.H. Shim (CEA-Grenoble), S. Bertaina (L2MP- Marseille), B. Malkin (Univ-Kazan).

Chemistry: A.M. Tkachuk (Univ-St. Petersbourg), T. Mitra (Univ-Bielefeld), A.Müller (Univ-Bielefeld).

Rabi oscillations of the 8 +1 electro-nuclear transitions of Er (0.001%):$CaWO_4$



Large anisotropy of Rabi frequencies



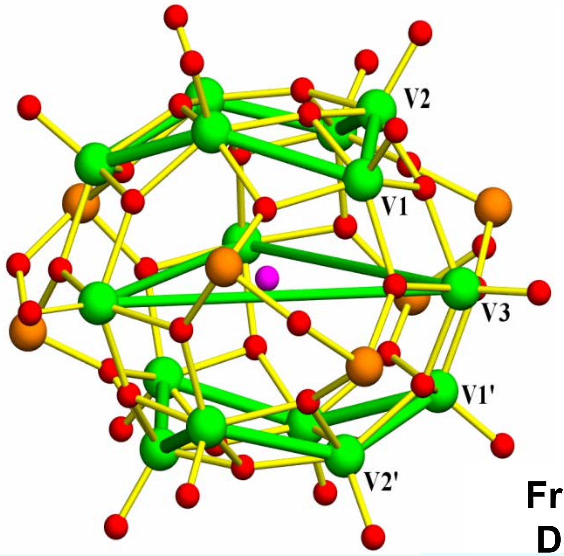
An effect of strong spin-orbit coupling → « **Spin-orbit qubits** »

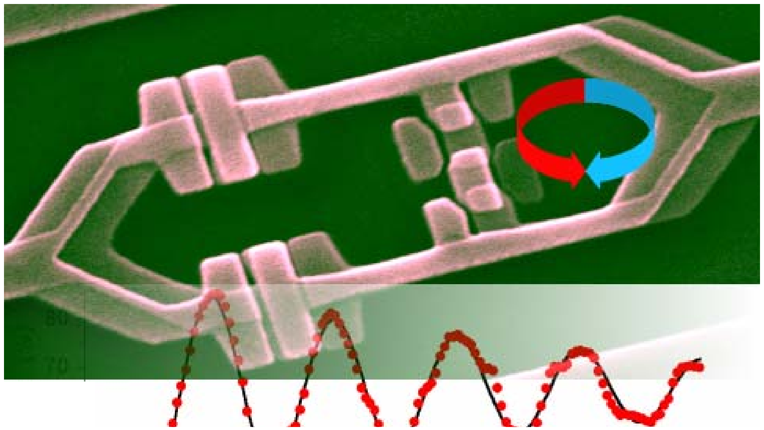
Phys. Rev. Lett. 24 Nov (2009)

CONCLUSION

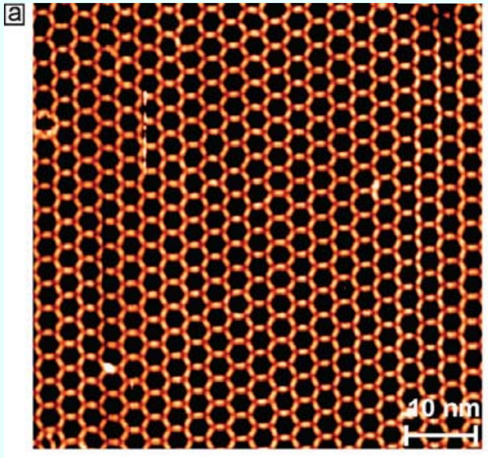
Entanglement of photons with complex molecules with huge Hilbert spaces

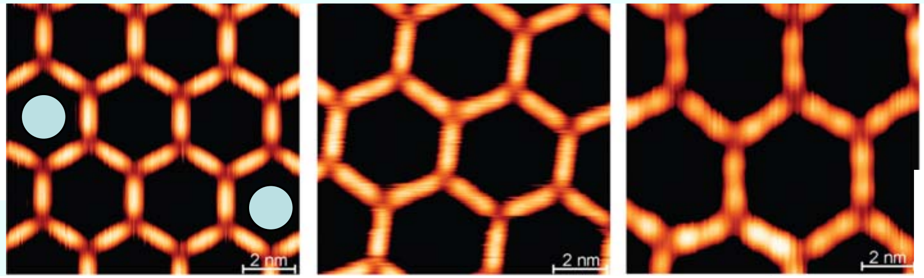
Self-organized 2D supra-molecular depositions become possible





From: I. Chiorescu, Y. Nakamura, K. Hartmans, H. Mooij et al,
Delft University of Technology





M. Ruben, J. V. Barth et. al., INT Karlsruhe, TU Munich

100 μs expected

Among the immediate projets: Rabi oscillations of a single molecule

1- Intoduction:

A Brief History of Mesoscopic Quantum Tunneling

70’s: Search for « macroscopic quantum tunnelling » phenomena
(… Schrödinger, Leggett)

- **1981** **First evidence of MQT in J - J** (R. Voss & R. Webb, IBM Yorktown-Heights)
- **1973 -1988** **Rare-earths with « narrow domain walls »:** Dy_3Al_2, $SmCo_{3.5}Cu_{1.5}$

  T-independent relaxation
- **80’s-90’s** **Films, nanoparticles ensembles**: a-SmCo, a-TbFe, $(TbCe)Fe_2$,…

  Theory: T. Egami R. Schilling, J.L. van Hemmen, P. Stamp, E. Chudnovsky, L. Gunther, N. Prokof’ev, …
- **90’s** **Two directions**: 1) single particle → **Micro-SQUIDs**

  2) ensembles of identical nanoparticles ↘

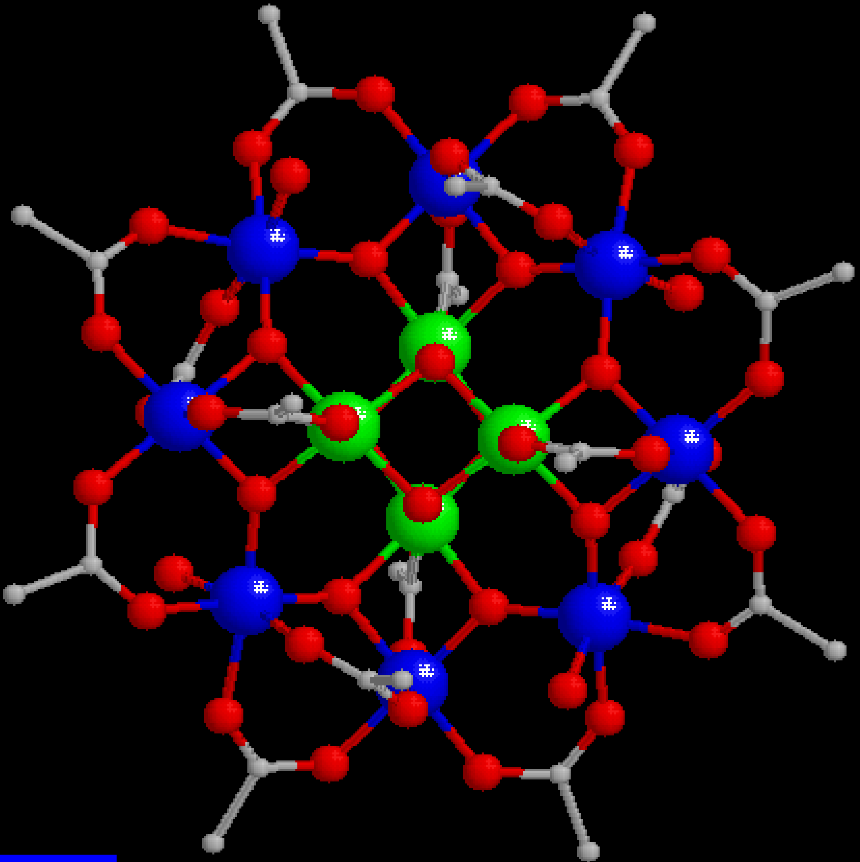
  Single Molecules Magnets

2- Incoherent quantum dynamics of Mn12-ac, RE-ions

🔵 Mn(III)
S=2

🟢 Mn(IV)
S=3/2

Total Spin
= 10



Mn12 acetate (very schematic)

T. Lis, Acta. Cryst. 1980

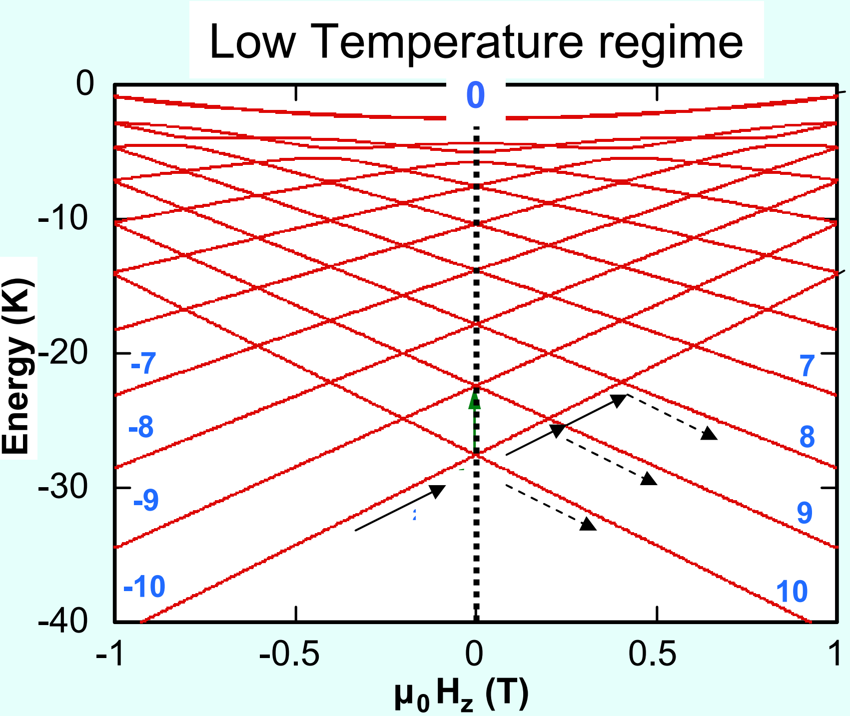
Single molecule magnets (Mn_{12}-ac)

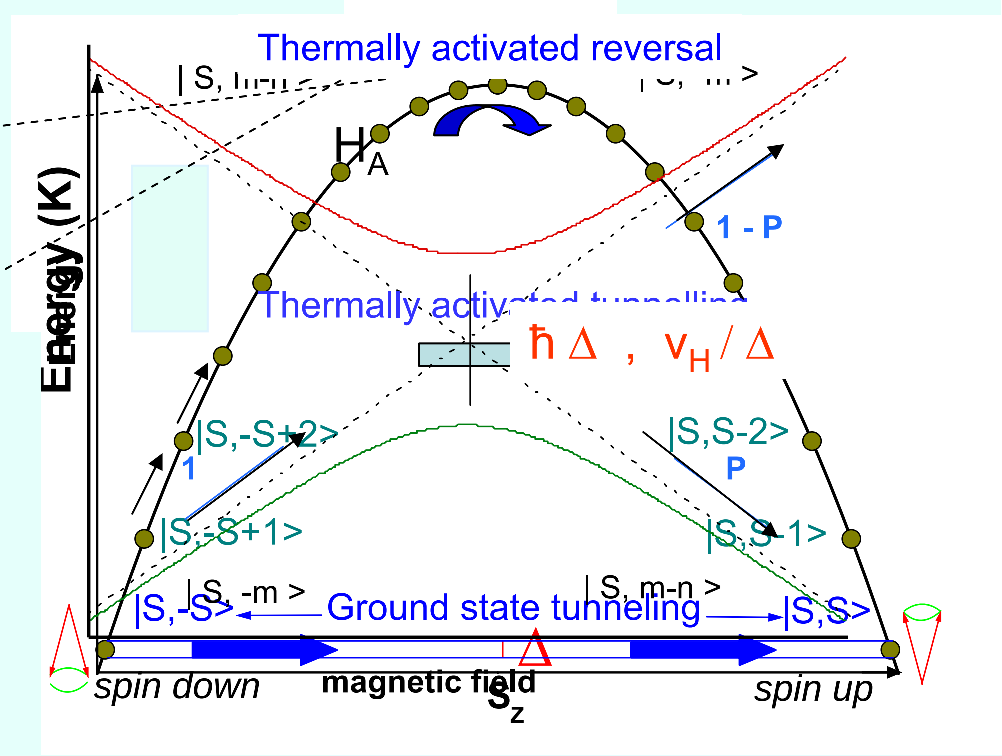
Macroscopic quantum magnet

From Kunio Awaga, Nagoya university

Classical barrier and tunnelling of a collective spin (S=10)

$$H = -DS_z^2 - BS_z^4 - g\mu_B S_z H_z - g\mu_B(S_+ + S_-)H_x/2 + E(S_+^2 + S_-^2) - C(S_+^4 + S_-^4)\ldots$$





H ≠ 0

S^+ Lanc

$$\Gamma \propto \Delta^2 \propto (TS^n/DS^2)^{4S/n}$$

H = 0

$$\Delta \propto (TS^n/DS^2)^{2S/n}, \quad n \le 2S$$

Probability:

$$P_{LZ} = 1 - \exp[-\pi(\Delta/\hbar)^2/\nu c] \approx \Delta^2/c$$

Resonances « under the barrier »

From quantum relaxation to coherence in magnetic systems

nanoparticles, single-molecule magnets, single-ions

B. Barbara
Institut Néel, CNRS, Grenoble

Introduction

Incoherent quantum dynamics of Mn_{12}-ac, RE-ions

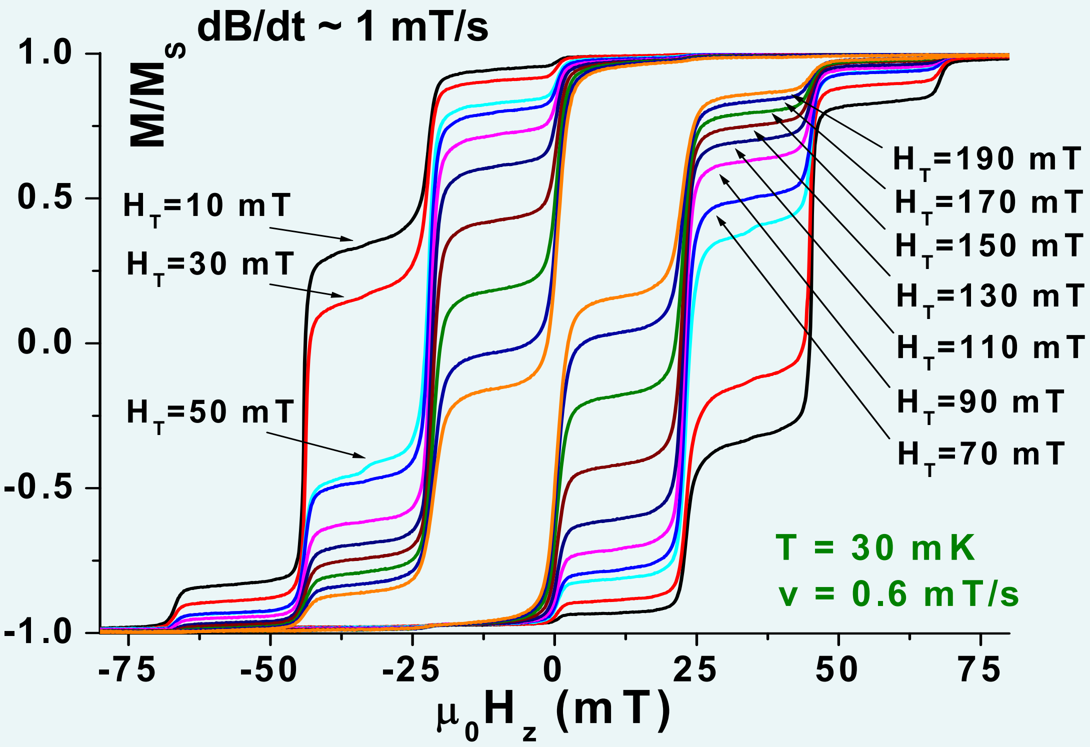
Coherent quantum dynamics of RE-ions
« The spin-orbit qubits »

Coherent quantum dynamics of the V_{15} SMMs
« Nanometer-size spin qubits »

Conclusion

Acceleration of quantum dynamics in a transverse field

Ho $^{3+}$ ions in $YLiF_4$



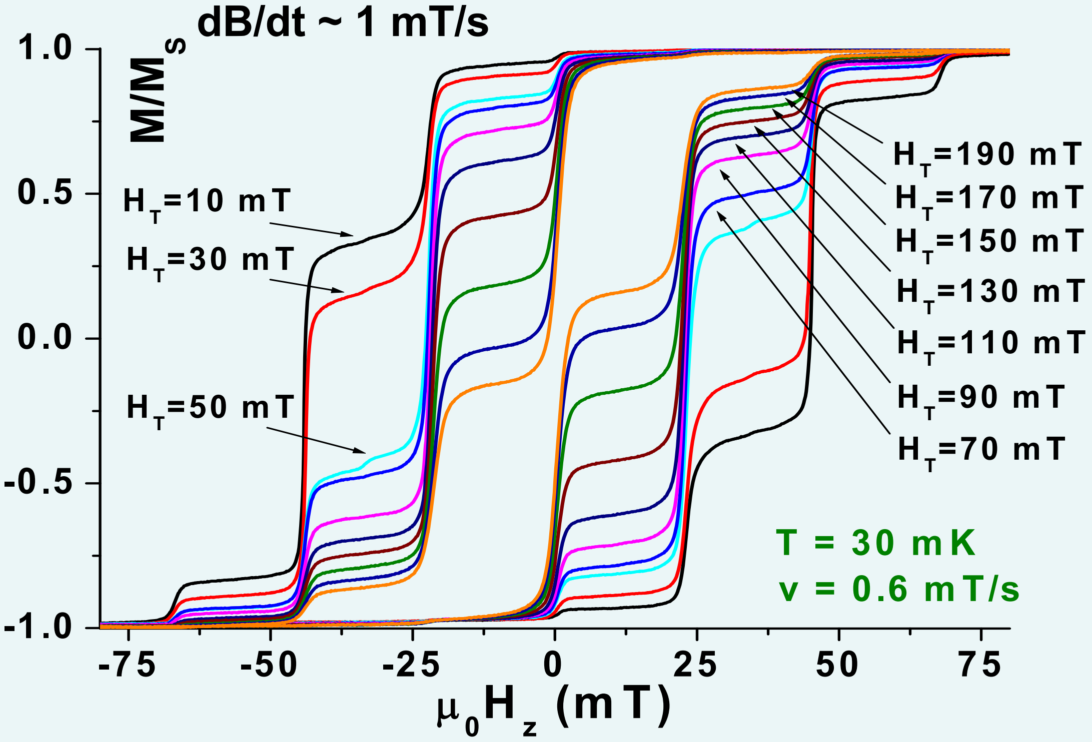
…. slow sweeping field: $\tau_{meas} >> \tau_{bott} > \tau_1$

Near thermodynamical equilibrium at the cryostat temperature…

Giraud et al, Phys. Rev. Lett. (2001)

Acceleration of quantum dynamics in a transverse field

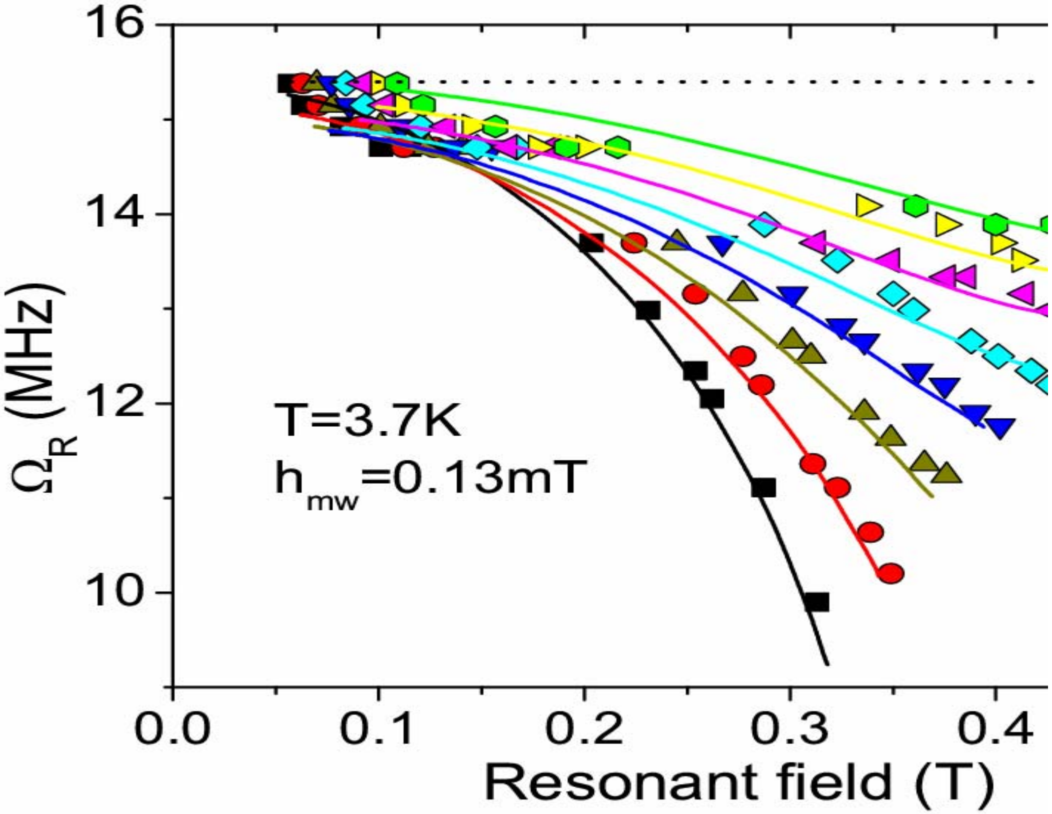
Ho $^{3+}$ ions in $YLiF_4$



…. slow sweeping field: $\tau_{meas} >> \tau_{bott} > \tau_1$

Near thermodynamical equilibrium at the cryostat temperature…

Giraud et al, Phys. Rev. Lett. (2001)

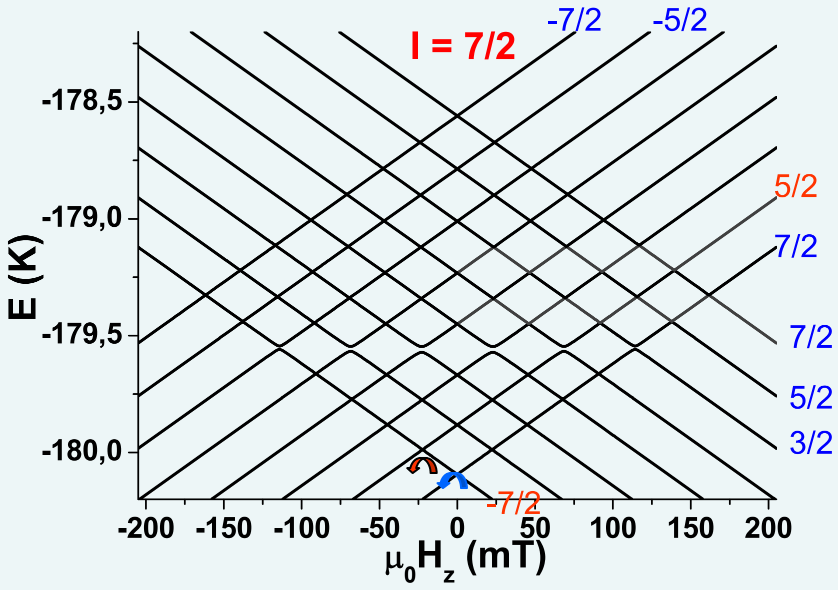

16
14
12
10
0.0
0.1
0.2
0.3
0.4
Ω_R (MHz)
Resonant field (T)
T=3.7K
h_{mw}=0.13mT

Ising CF ground-state + hyperfine Interactions

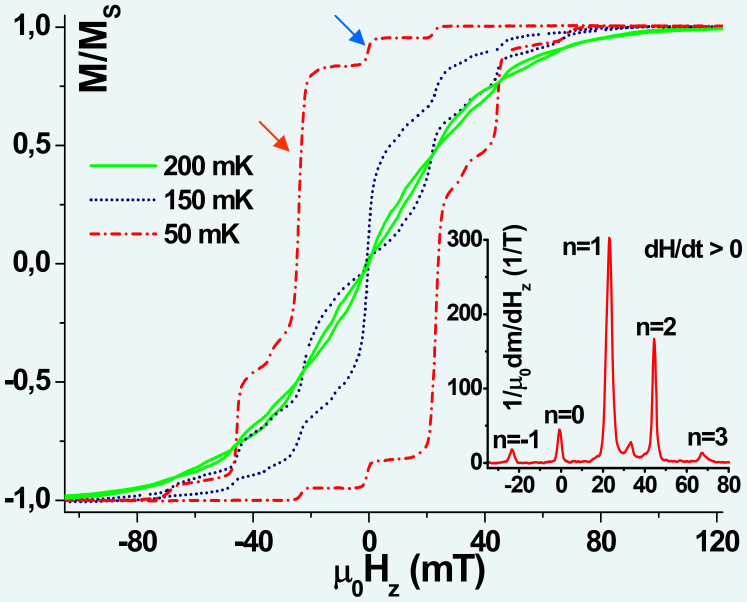
$$H = H_{CF} + A_J (J^+ I^- + J^- I^+)/2$$

$$H_{CF} = \alpha_J B_2^0 O_2^0 + \beta_J (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma_J (B_6^0 O_6^0 + B_6^4 O_6^4)$$

The ground-state doublet ⟹ 2(2 x 7/2 + 1) = 16 states



$g_J \mu_B H_n = n.A/2$



A = 38.6 mK

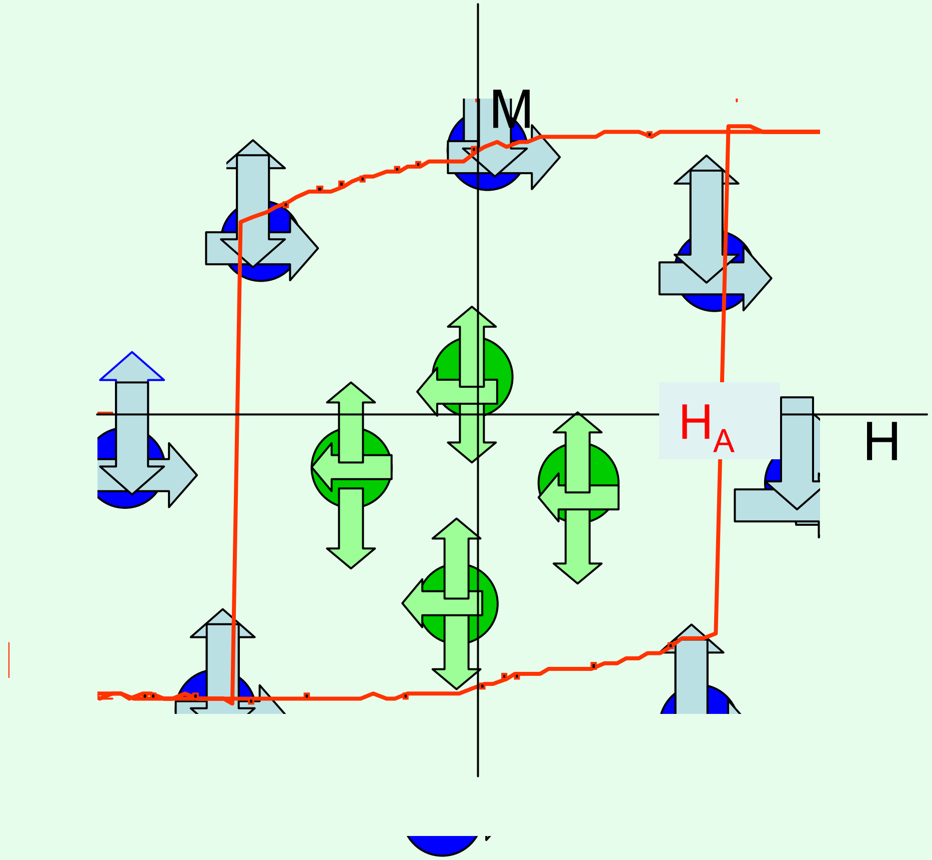
Co-Tunneling of electronic and nuclear momenta

Giraud et al, Phys. Rev. Lett. (2001)

Mn12 acetate (very schematic)

● Mn(III)
S=2

● Mn(IV)
S=3/2



Total Spin
= 10

Interferences

1920 – Davisson and Germer (electron bounces off a chunk of nickel)

1961 – Jönsson (single electrons double slit)

1974 – Merli (one electron at the time)

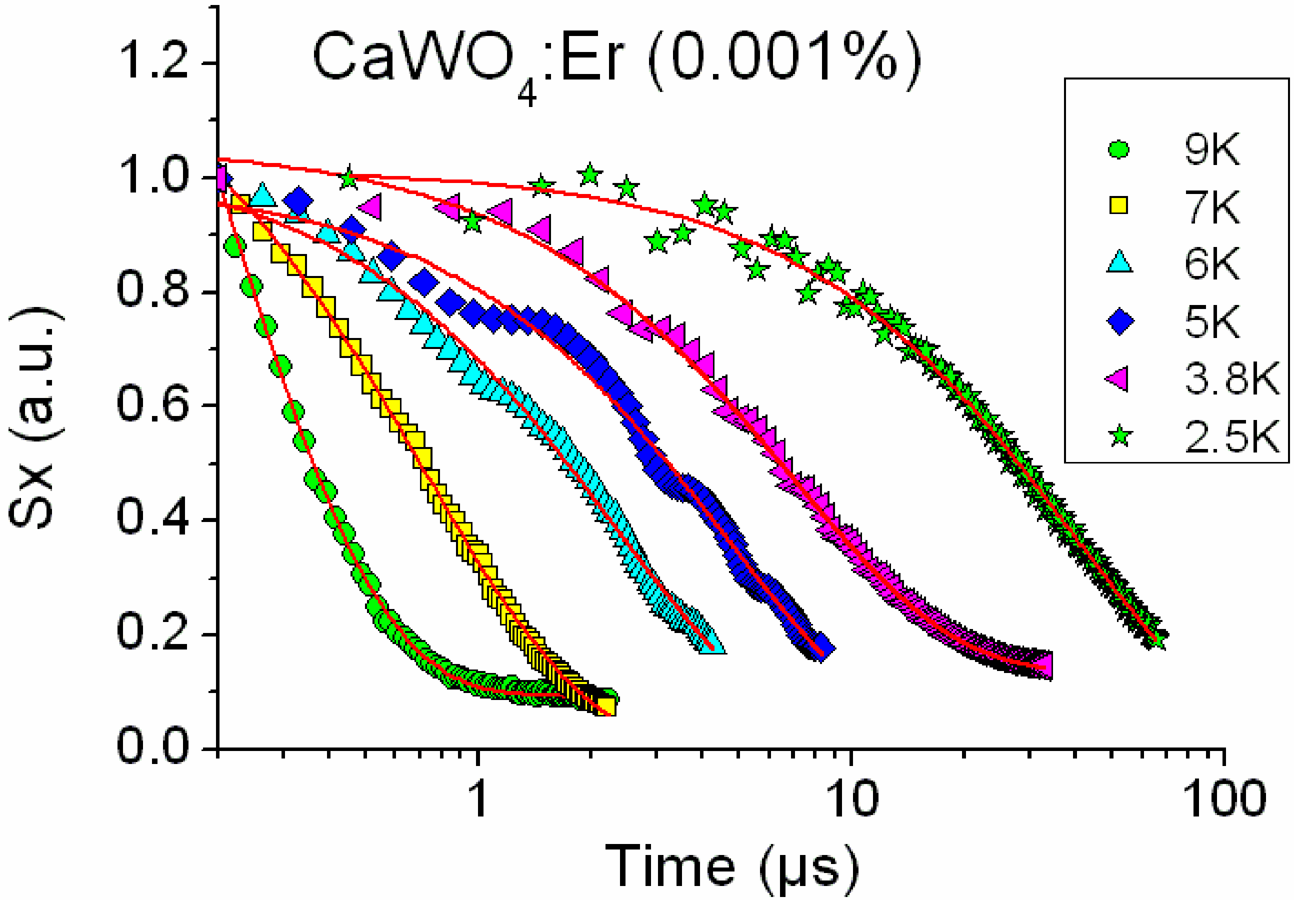
1989 – Tonomura (one electron at the time)

Decoherence

1970 – Zeh
1980 – Zurek… Giulini, Schlosshauer …..
1981 – Leggett, Caldeira (quantum dissipation and quant → class transition)
1996 – Stamp, Prof'ev (spin-bath)

….

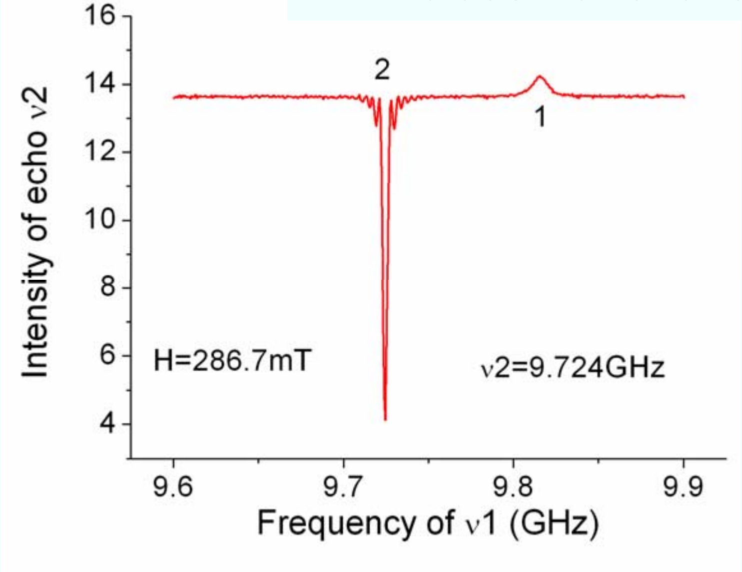
Coherence times T_2 vs T

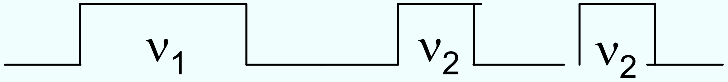


→ T_2 = 1ms, 4K

Coherent multilevel manipulations in Gd:CaWO$_4$

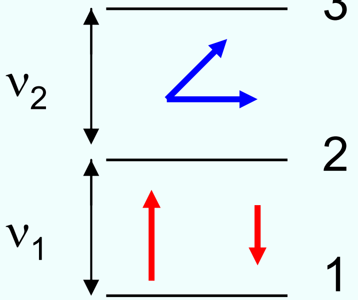
2 close transtions: 1st excite and 2nd probe

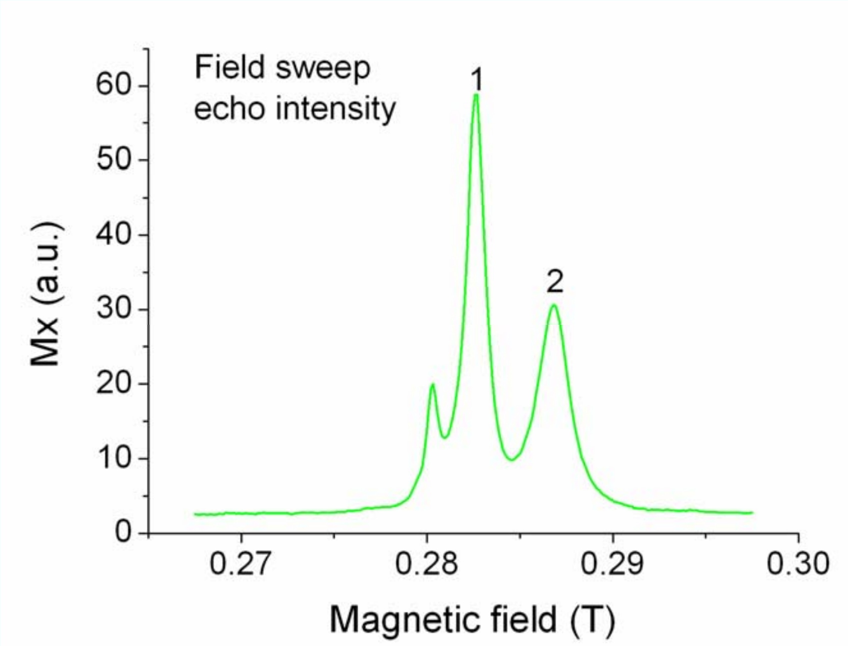


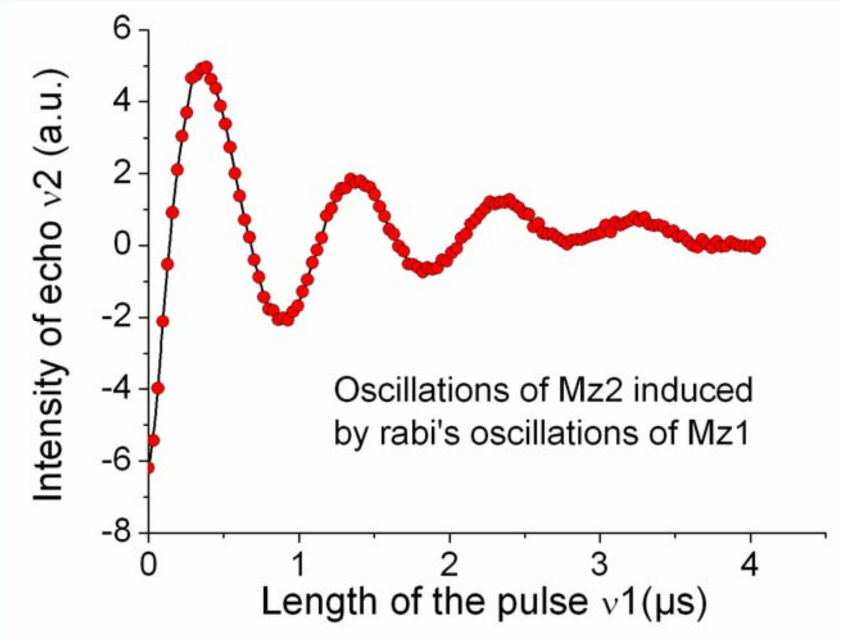


τ_2 ≈ 1ms at 4K

≈ 10 μs at 80 K



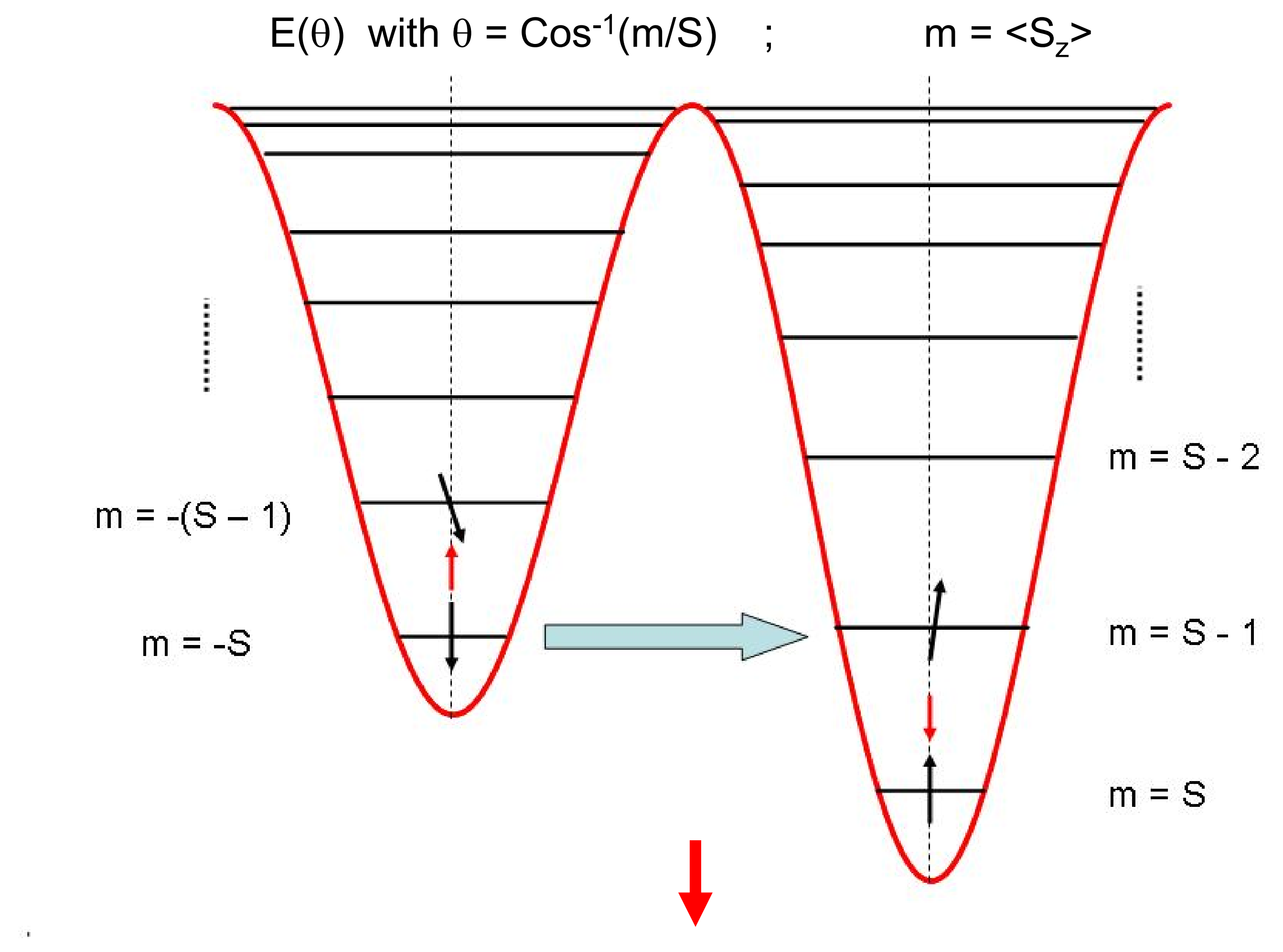




Basic interpretation

$H_D = -DS_z^2 \ldots - g\mu_B S_z H_z$

$H_{ND} = B(S_x^2 + S_y^2)\ldots$

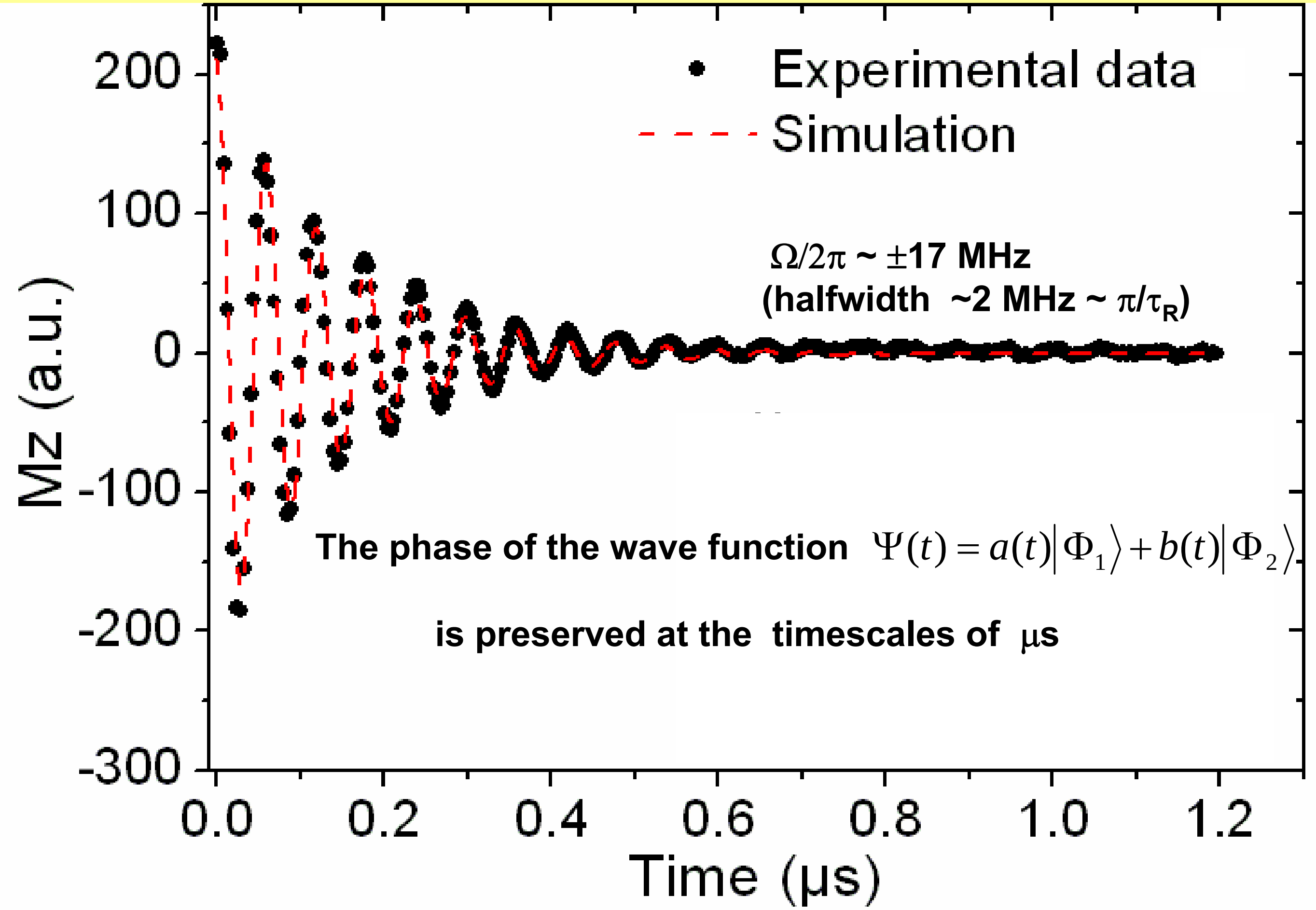


$E(\theta)$ with $\theta = \mathrm{Cos}^{-1}(m/S)$; $m = \langle S_z \rangle$

Quantum relaxation

An example of E-N Rabi oscillations

Er(0.001%):CaWO$_4$ (H=0.522 T // c, h=0.15 mT // b, T=3.5 K)



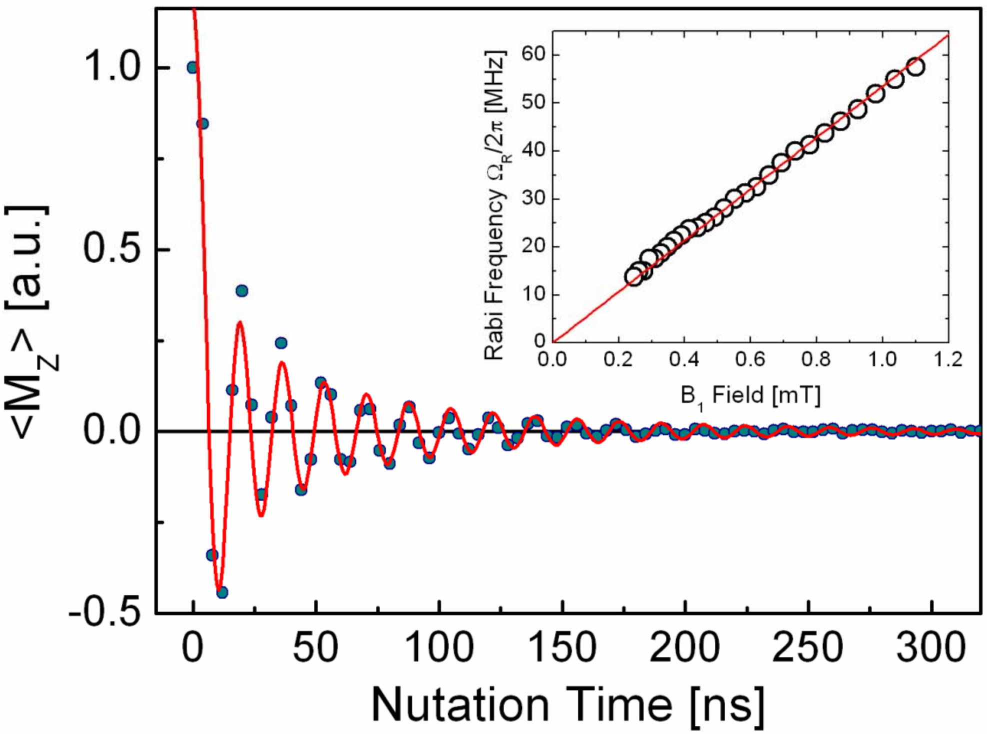
$\Omega/2\pi$ ~ ±17 MHz
(halfwidth ~2 MHz ~ π/τ_R)

The phase of the wave function $\Psi(t) = a(t)|\Phi_1\rangle + b(t)|\Phi_2\rangle$

is preserved at the timescales of μs

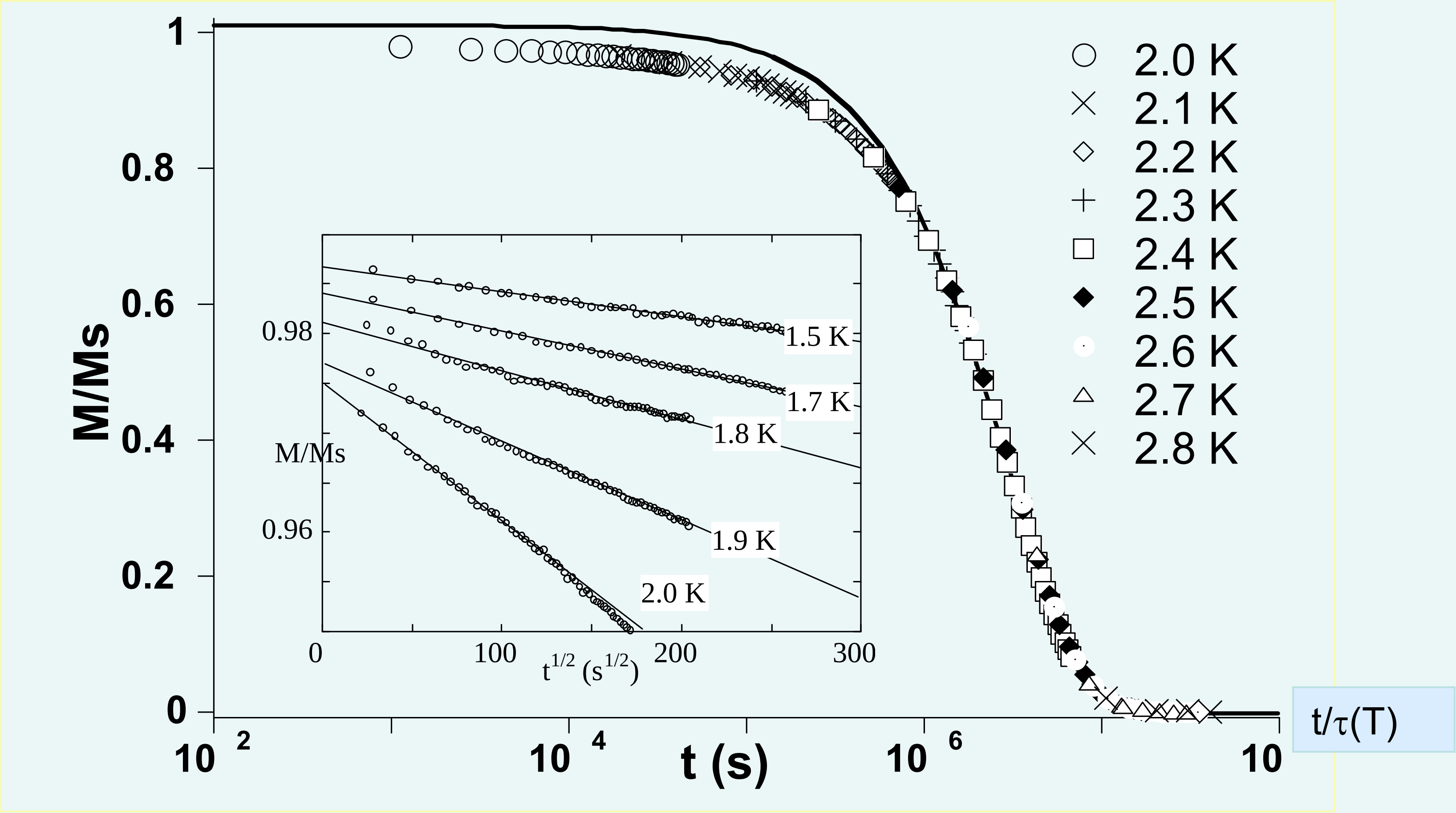
τ_R= 0.2 μs << τ_2 ~ 7μs

S. Bertaina et al Nature nanotechnology, Jan. 2007


1.0
0.5
0.0
-0.5
<M_Z> [a.u.]
0
50
100
150
200
250
300
Nutation Time [ns]
Rabi Frequency Ω_R/2π [MHz]
0
10
20
30
40
50
60
0.0
0.2
0.4
0.6
0.8
1.0
1.2
B_1 Field [mT]

Cross-over from exponential to square-root relaxation

(Predicted by Prokofiev and Stamp, PRL 80, 5794, 1998)



L. Thomas, A. Caneschi and B. Barbara
J. Low Temp. Phys. (1998) and Phys. Rev. Lett. (1999).

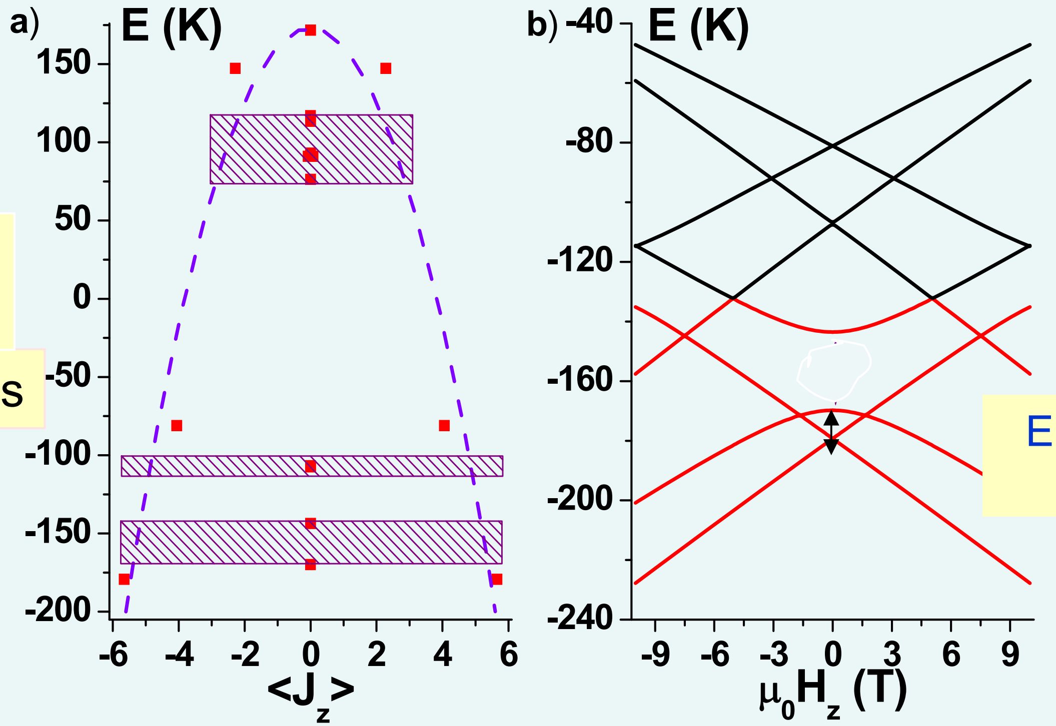
CF levels and energy barrier of Ho^{3+} in $LiYF_4$:Ho

$$H_{CF} = \alpha_J B_2^0 O_2^0 + \beta_J (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma_J (B_6^0 O_6^0 + B_6^4 O_6^4)$$

Strong mixing

⇩

Barrier short-cuts



Energy barrier

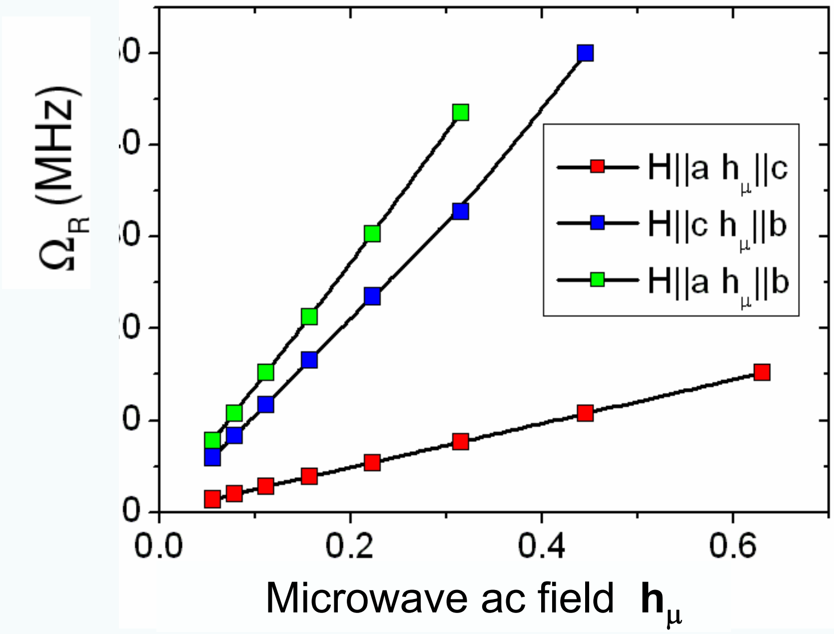
(~ 10 K)

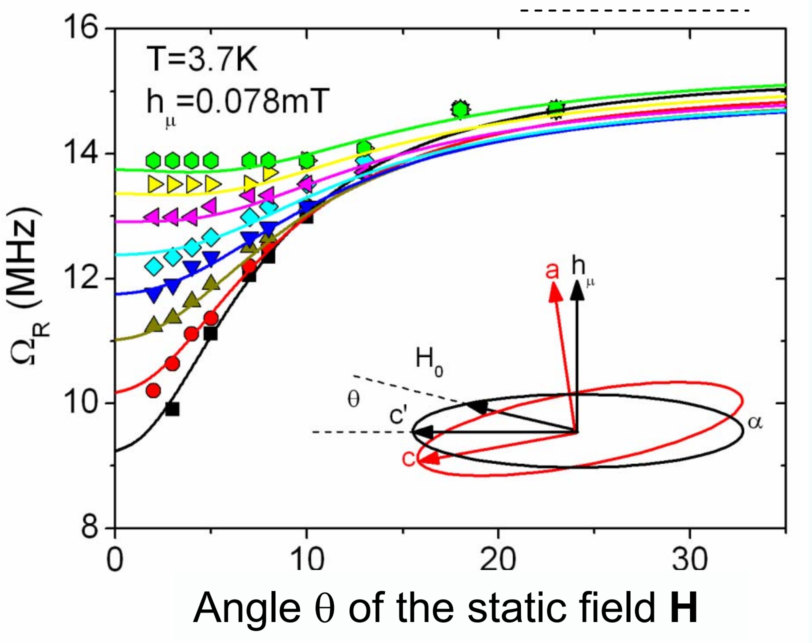
B20 = 0.606 K, B40 = -3.253 mK, B44 =- 42.92 mK, B60 =-8.41mK, B64 =- 817.3mK

Sh. Gifeisman et al, Opt. Spect. (USSR) 44, 68 (1978); N.I. Agladze et al, PRL, 66, 477 (1991)

Experimental evidence of anisotropic Rabi frequency

Measurements and fits (diagonalisation of the the C-F electro-nuclear Hamiltonian





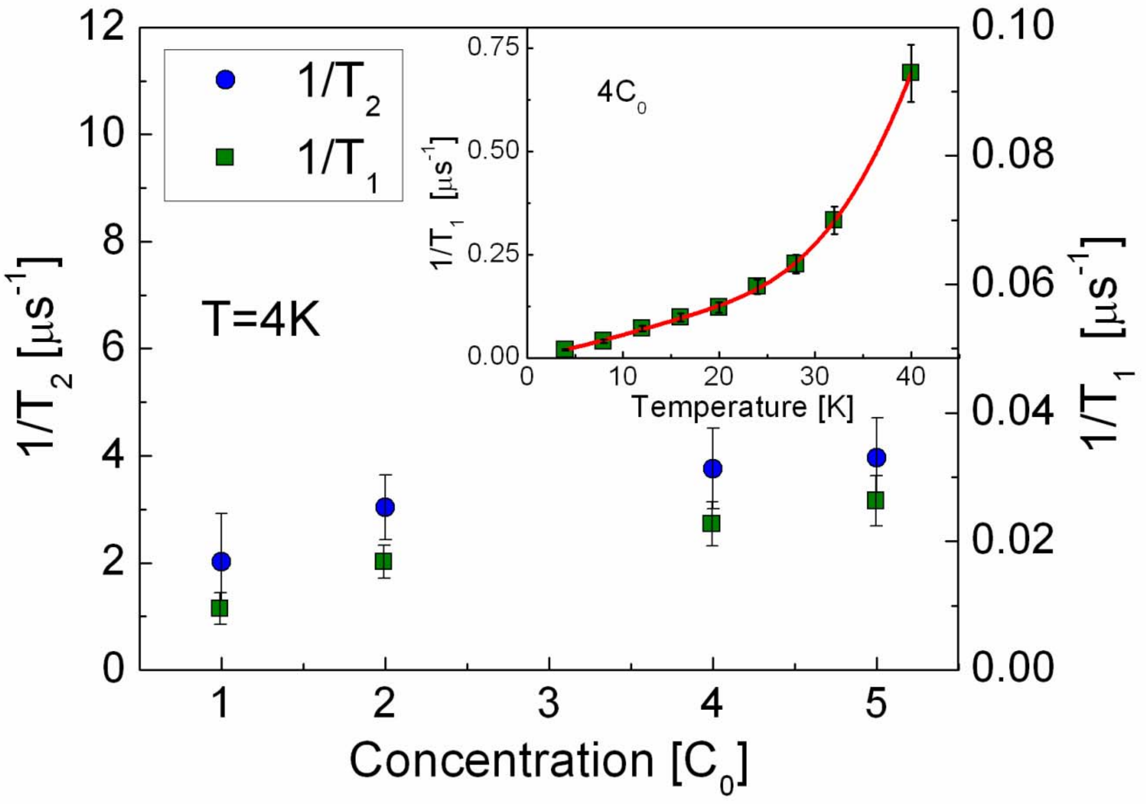
Rabi frequency: $\hbar\Omega_R = g_J \mu_B h_\mu \langle \varphi_1 | J_\mu | \varphi_2 \rangle$

$\hbar\Omega_R = g_J \mu_B \vec{J}_{1,2} \vec{h}$ where $\vec{J}_{1,2} = \left\| \langle \varphi_1(\vec{H}) | \vec{J} | \varphi_2(\vec{H}) \rangle \right\|$

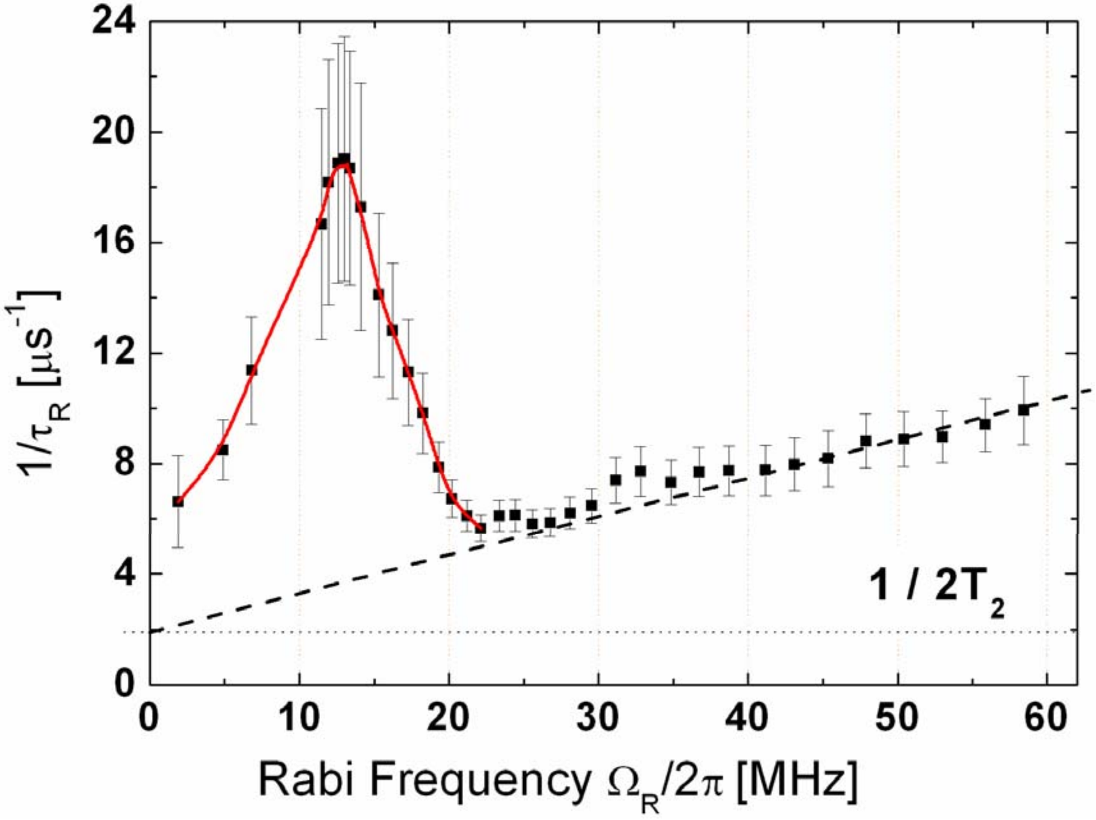
The Rabi frequency changes in accordance with the local symmetry

An effect of strong spin-orbit coupling → « **Spin-orbit qubits** »

Phys. Rev. Lett. 6 Feb (2009)



1/T$_2$ [μs^{-1}]
1/T$_1$ [μs^{-1}]
Concentration [C$_0$]
0
2
4
6
8
10
12
0.00
0.02
0.04
0.06
0.08
0.10
1
2
3
4
5
● 1/T$_2$
■ 1/T$_1$
T=4K
4C$_0$
1/T$_1$ [μs^{-1}]
0.00
0.25
0.50
0.75
0
10
20
30
40
Temperature [K]



$1/\tau_R$ [μs^{-1}]
0
4
8
12
16
20
24
0
10
20
30
40
50
60
Rabi Frequency $\Omega_R/2\pi$ [MHz]
$1 / 2T_2$

Rare-earth ions

Mesoscopic physics of domain walls in single crystals

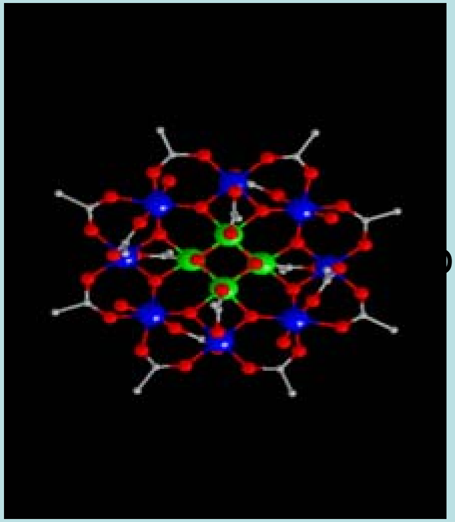
2001 1993 -1996 1986 - 1995 1973 - 1986

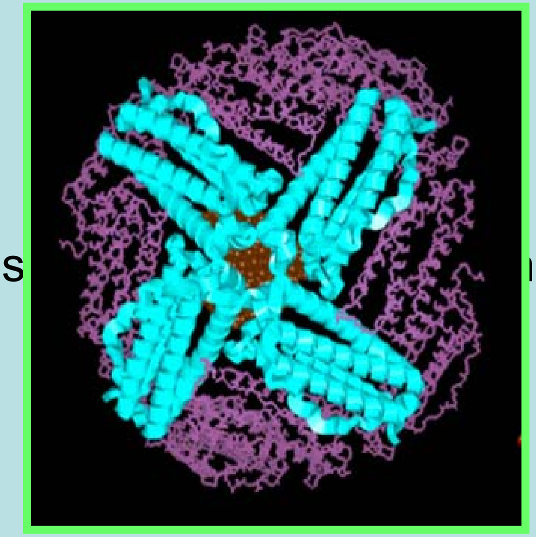
Single Molecule

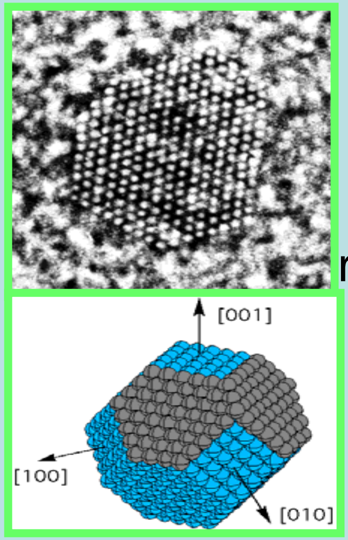
Magnetic Protein

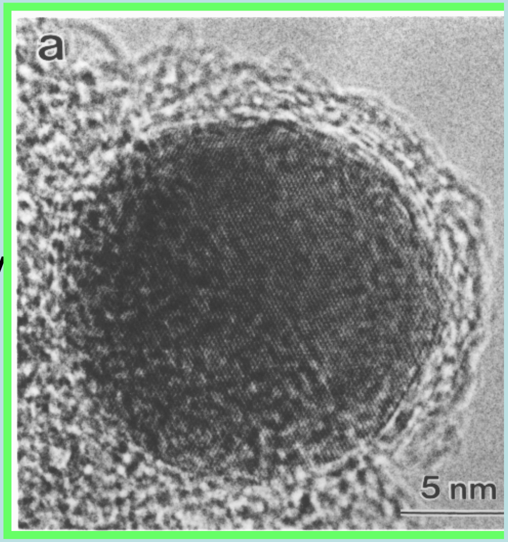
Cluster

Nanoparticle









1 nm

2 nm

3 nm

20 nm

For a short historical review, see:

K. Ziemelis, Nature, « Milestones on Spin », S19, March 2008
(Produced by Nature Physics)

Calculation of anisotropic Rabi frequencies

Local frame

Space product

$$H = H_{CF} + A_J \vec{I}\vec{J} - g_J \mu_B \vec{J}\vec{H}$$

$$\left|L, S, J, m_J\right\rangle \otimes \left|I, m_I\right\rangle$$

$$\Omega_R = g_J \mu_B \left\langle \phi_1(\vec{H}) \middle| \vec{J} \middle| \phi_2(\vec{H}) \right\rangle \vec{h}_{mw} / 2h$$

Rotating frame approximation

Truncation to lowest CF doublet

(anisotropic *g*-factor: $g_c \sim 1.25$ $g_{a\text{-}b} \sim 8.38$)

$$H(t) = H - g_J \mu_B \vec{J}.\vec{h}_{mw} \cos(\omega t)$$

$$\left|1/2, m_I\right\rangle \qquad \left|-1/2, m_I\right\rangle$$

↓

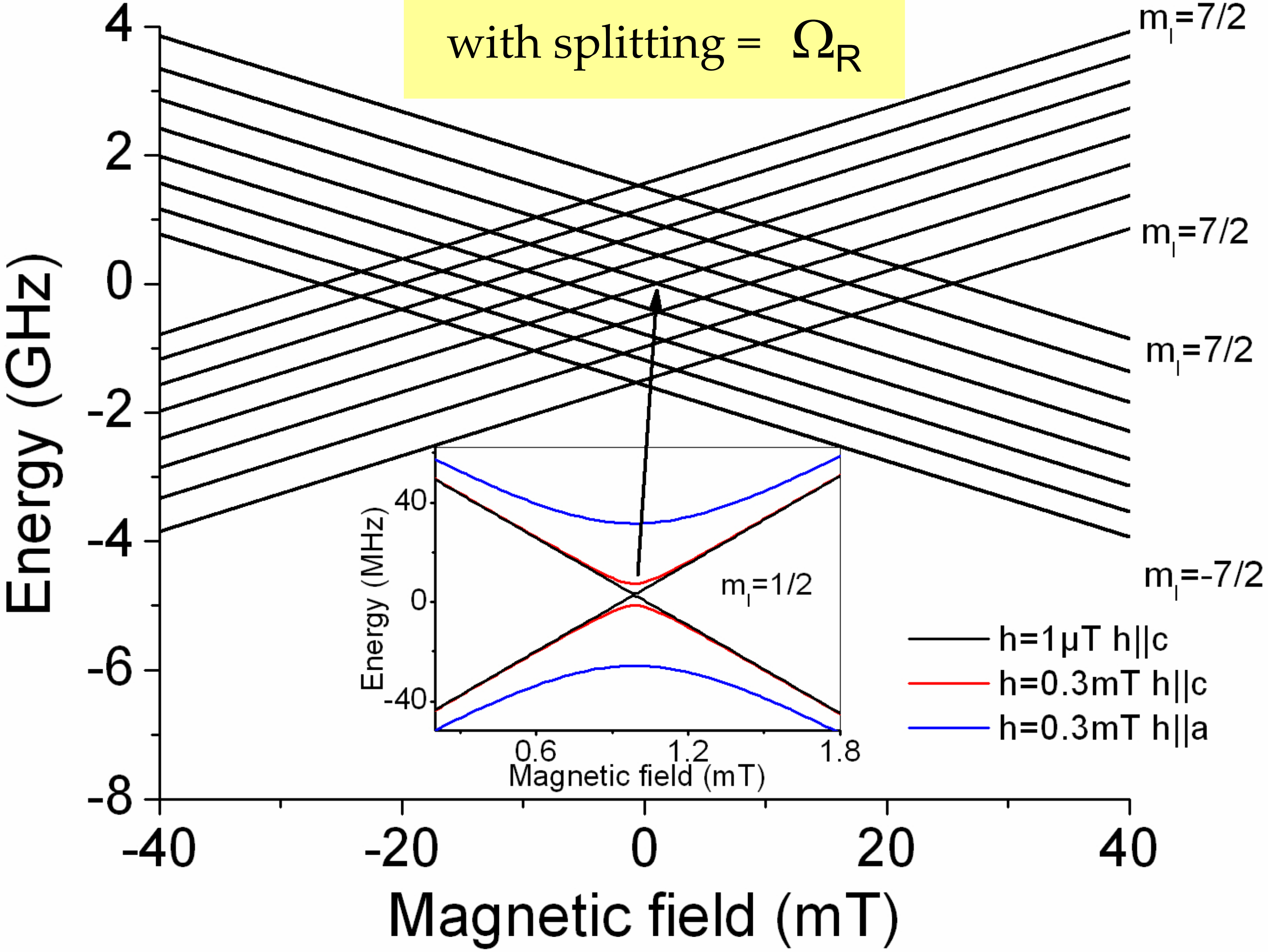
Time-independent Hamiltonian

*B*02=231cm^{-1}, *B*04=- 90cm^{-1}, *B*44=852 cm^{-1}, *B*06=- 0.6cm^{-1}, *B*46=396cm^{-1}
A = - 4.15 10^{-3} cm^{-1}

Rotating frame energy spectrum

The different states $\left|-1/2, m_I\right\rangle$ and $\left|1/2, m_I\right\rangle$ form avoided L.C.

with splitting = Ω_R



Bertaina *et al*, *Phys. Rev. Lett*. (2009)

Analytical calculations in the LFA

Coll. Boris Malkin, Kazan university

I=0 isotope

$$H = \mu_B\{g_{eff}(\theta)BS_{z'} + \frac{1}{2}(e^{i\omega t} + e^{-i\omega t})[h\sin\theta\cos\theta\frac{g_\perp^2 - g_\parallel^2}{g_{eff}(\theta)}S_{z'} + h\frac{g_\perp g_\parallel}{g_{eff}(\theta)}S_{x'} + g_\perp h_y S_{y'}]\}.$$

$$\Omega_R(\theta) = \frac{\mu_B g_\perp}{2\hbar}[\left(\frac{hg_\parallel}{g_{eff}(\theta)}\right)^2 + h_y^2]^{1/2}$$

$$g_{eff}(\theta) = (g_\parallel^2\cos\theta^2 + g_\perp^2\sin\theta^2)^{1/2}$$

I≠0 isotope

$$\Omega_R^{(m)}(\theta) = \frac{\mu_B g_\perp}{2\hbar}[\left(\frac{hg_\parallel}{g_{eff}(\theta)}[1 - \Delta_m(\theta)]\right)^2 + h_y^2]^{1/2}$$

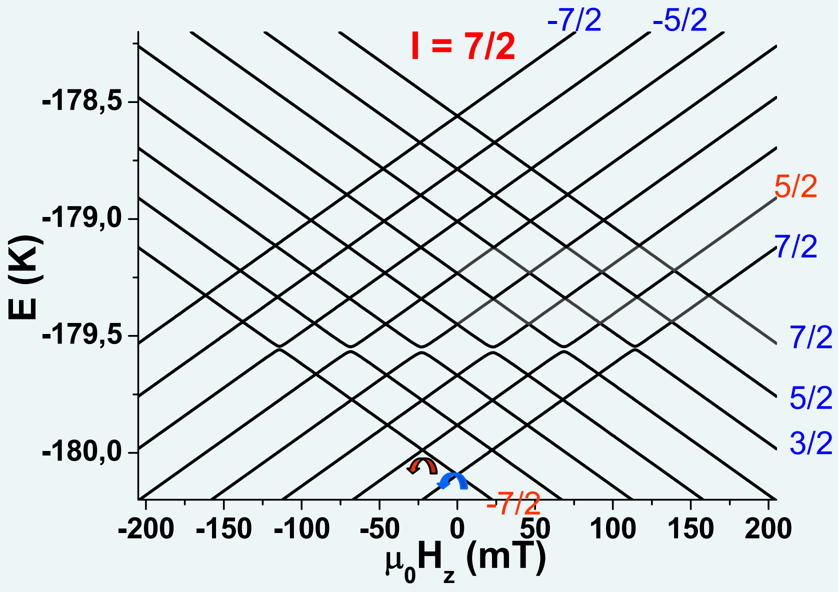
$$\Delta_m(\theta) = \frac{mA}{g_J g_{eff}(\theta)\hbar\omega_m}\frac{(g_\parallel^2 - g_\perp^2)^2(\sin\theta)^2(\cos\theta)^2}{g_{eff}^{(2)}(\theta)}$$

CF ground-state + hyperfine Interactions

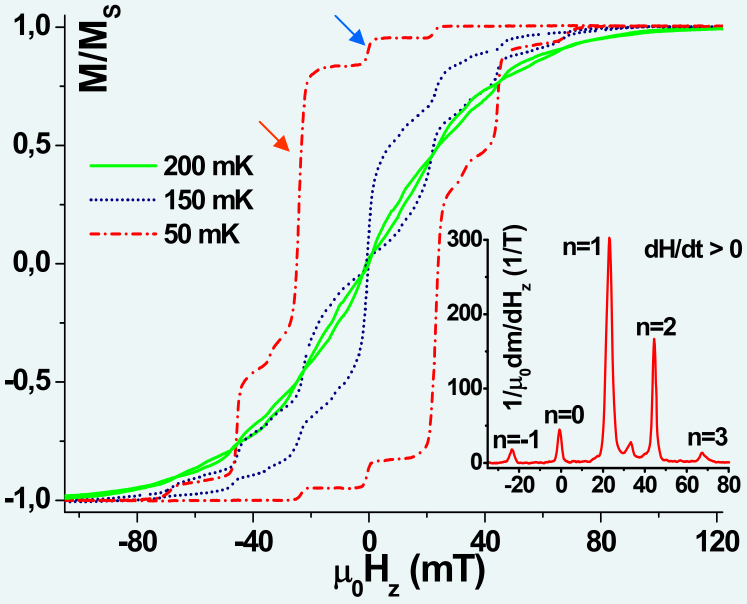
$$H = H_{CF} + A_J(J^+I^- + J^-I^+)/2$$

$$H_{CF} = \alpha_J B_2^0 O_2^0 + \beta_J (B_4^0 O_4^0 + B_4^4 O_4^4) + \gamma_J (B_6^0 O_6^0 + B_6^4 O_6^4)$$

The ground-state doublet ⟹ 2(2 x 7/2 + 1) = 16 states



$g_J \mu_B H_n = n.A/2$



A = 38.6 mK

Co-Tunneling of electronic and nuclear momenta

Phys. Rev. Lett. (2001, 2003)

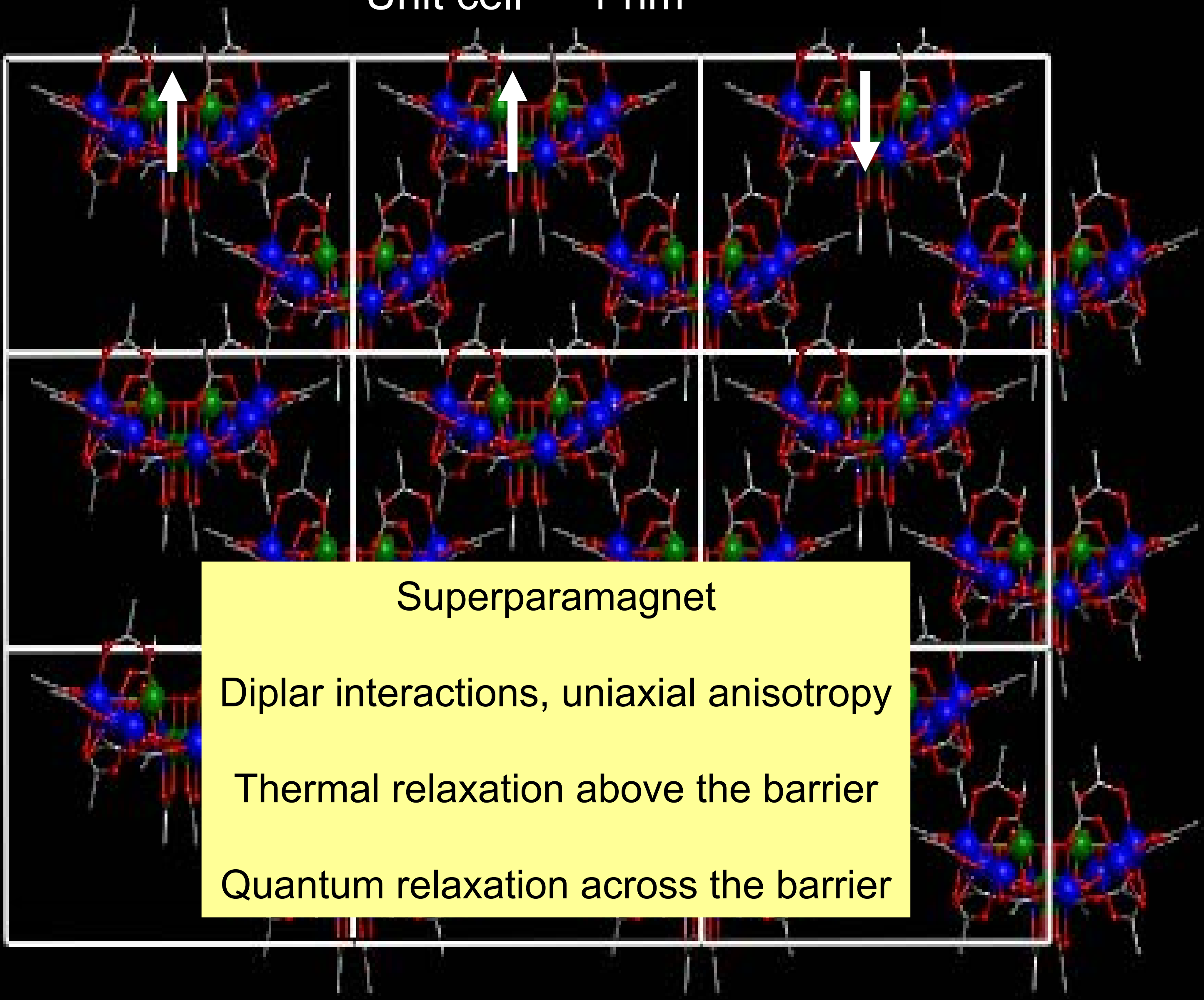
Single molecule magnets (Mn_{12}-ac)

Macroscopic quantum magnet

From Kunio Awaga, Nagoya university

Typical structure of a single molecule magnet

Unit cell ~ 1 nm



Superparamagnet

Diplar interactions, uniaxial anisotropy

Thermal relaxation above the barrier

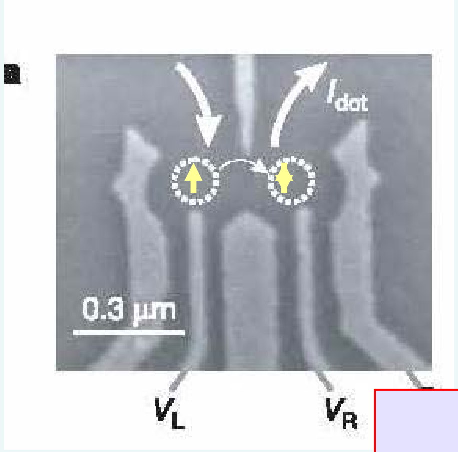
Quantum relaxation across the barrier

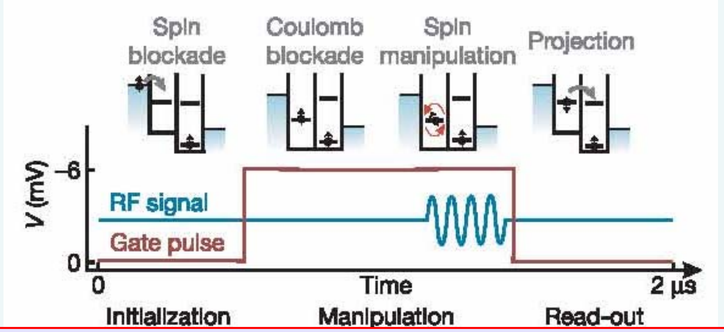
Driven oscillations of a single electron spin in a quantum dot

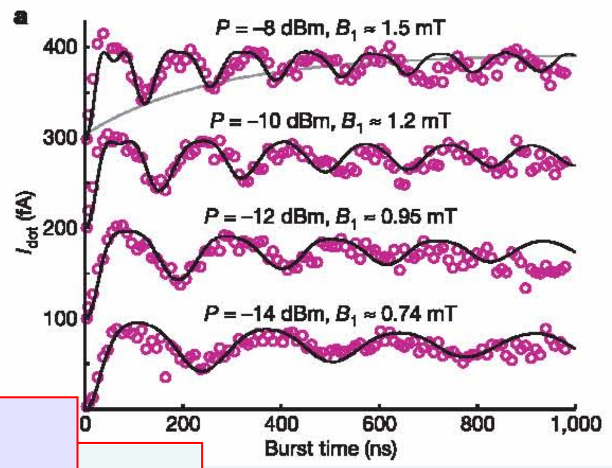
F. Koppens, C. Buizert, K.J. Tielrooj, I.T. Vink, K.C. Nowack, T. Meunier, L.P. Kowenhoven, L. Vandersypen

Nature, 17 Aug., 2006

Singlet / triplet states entangle with nuclear spin states ($\Delta \approx \sigma$)







Coherence limitations ?

$B_{mw} \sim B_{N\text{-}S}$ → N-S affects spin manipulations

Low fidelity

to s

$T_{S\text{-}B}$ >> T_{Rea}

Nuclear S-B is froze
each measureme

$T_{S\text{-}B}$ << T_{Int}

Average over distributed Ramsey frequencies

Broad distribution of Larmor frequencies →
Destructive additions, $T_{2*} \sim h/\sigma$

Rabi oscillations
(driven oscillations)
$T_R \approx 1$ µs

Spin-echo
(also affectd by NS)
$T_{S\text{-}E} \approx 1$ µs

Ramsey oscillations
(free precession decay)
$T_{Rm} \approx 30$ ns

Distribution of Ω_R limited by inhomogeneous level broadening ε.
Addition of $\Omega_{R0} < \Omega_R < (\Omega_{R0} + \varepsilon 2)1/2$

Suppresses fast NS

fluctuations