LABORATOIRE DE SPECTROCHIMIE
INFRAROUGE ET RAMAN
UMR 8516

Coherence lifetimes of Zeeman and hyperfine transitions in Nd:YSO measured by ESR spectroscopy

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AGENDA

- Introduction
- CW ESR
- Coherence lifetimes of Zeeman transitions
- Coherence lifetime of hyperfine transitions
- Spectral diffusion

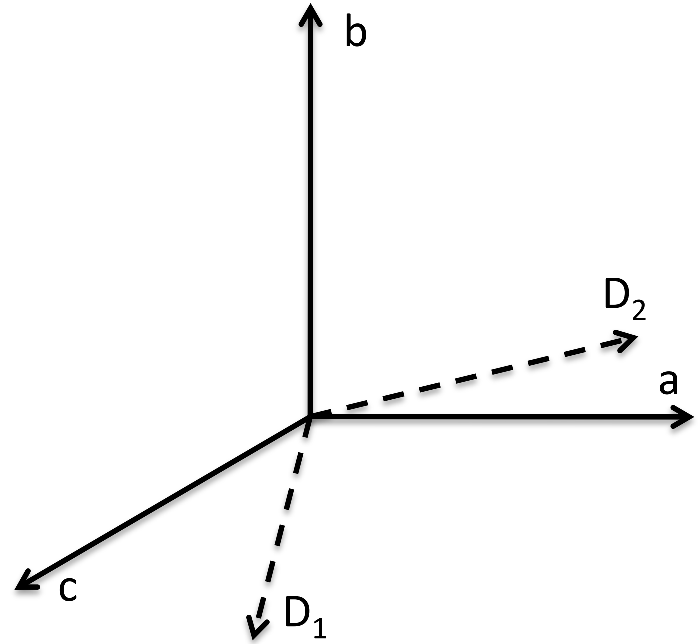
INTRODUCTION

AN INTRODUCTION TO Nd:YSO

Space group $P2_1/c$: monoclinic
2 main sites :

Site 1 : 6 + 1 oxygens R =[2.16 – 2.34; 2.61Å]
Böttger, Afzelius
Site 2 :   6 oxygens R =[2.16 – 2.26Å]

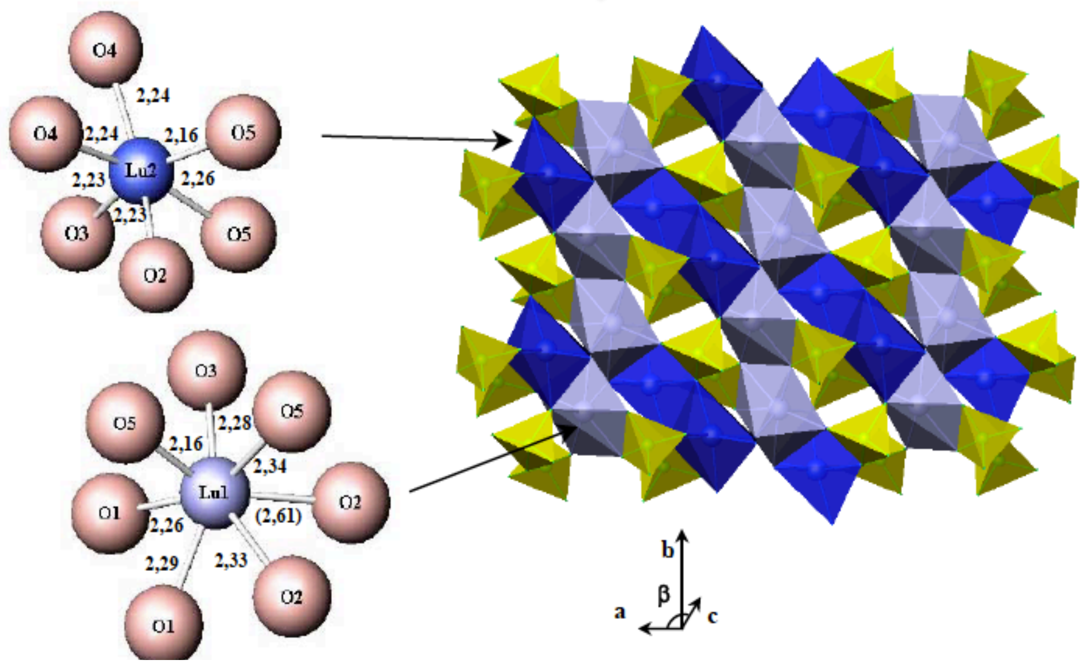
Due to its size, Nd^{3+} mainly substitutes in site 1



Y^{3+}	Nd^{3+}	Er^{3+}
0.892	0.995	0.881

^{142}Nd 27.2%
^{143}Nd 12.2%
^{144}Nd 23.8%
^{145}Nd 8.3%
^{146}Nd 17.2%
^{148}Nd 5.7%
^{150}Nd 5.6%

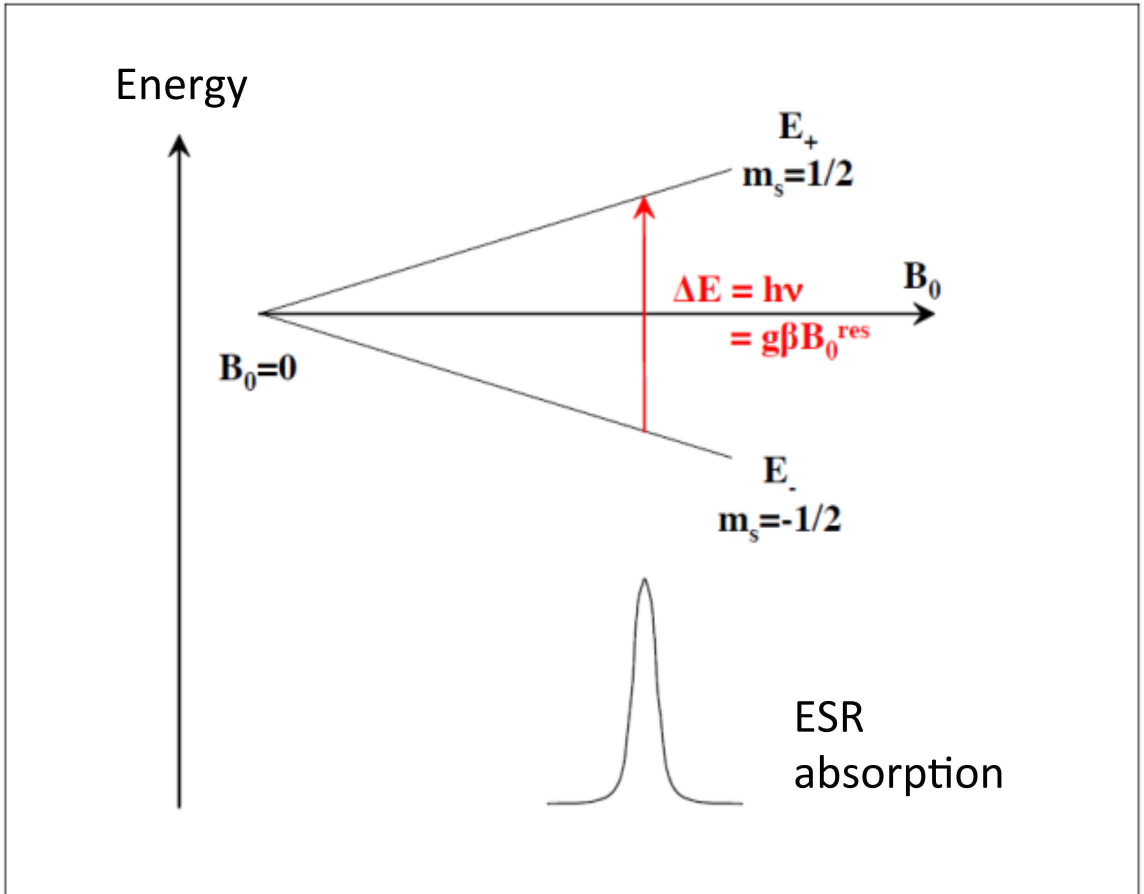
Both Zeeman and hyperfine transitions

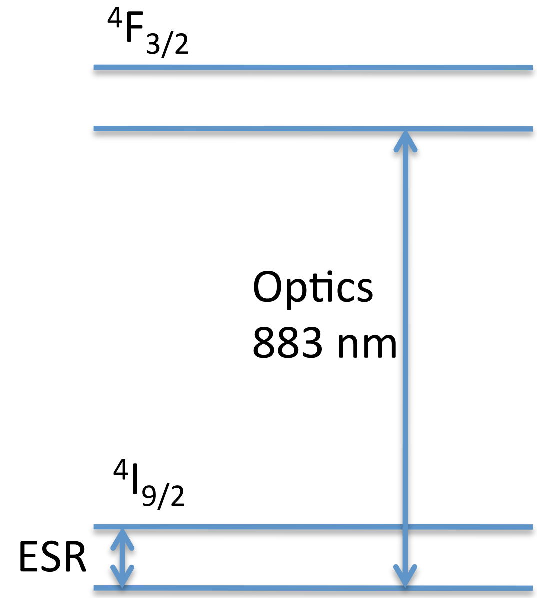


L. Pidol, thèse de doctorat, UPMC, 2004

CW ESR

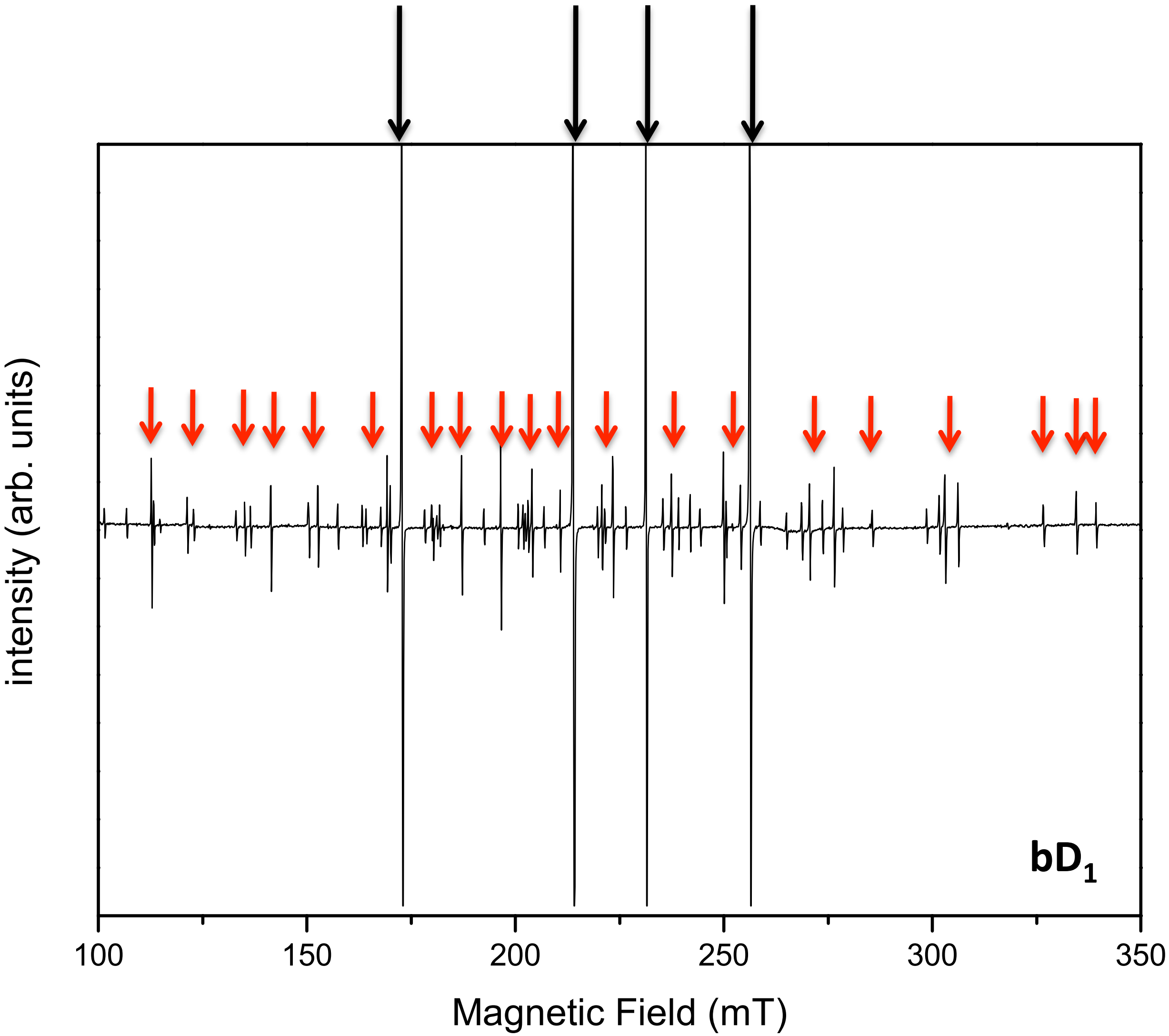
ESR SPECTROSCOPY

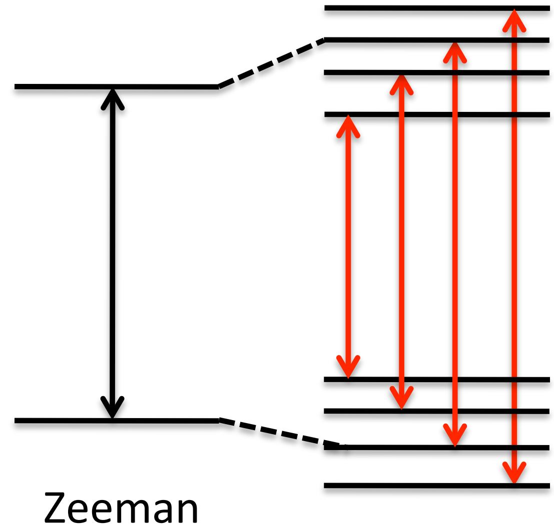




X band ESR
Freq. : 9.4 (CW)
9.8 GHz (Pulsed)
Cavity mode
B : 0 – 10000 Gauss
Power: 0.01 mW
The derivative
is recorded

ESR SPECTROSCOPY





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^{148}Nd 5.7%
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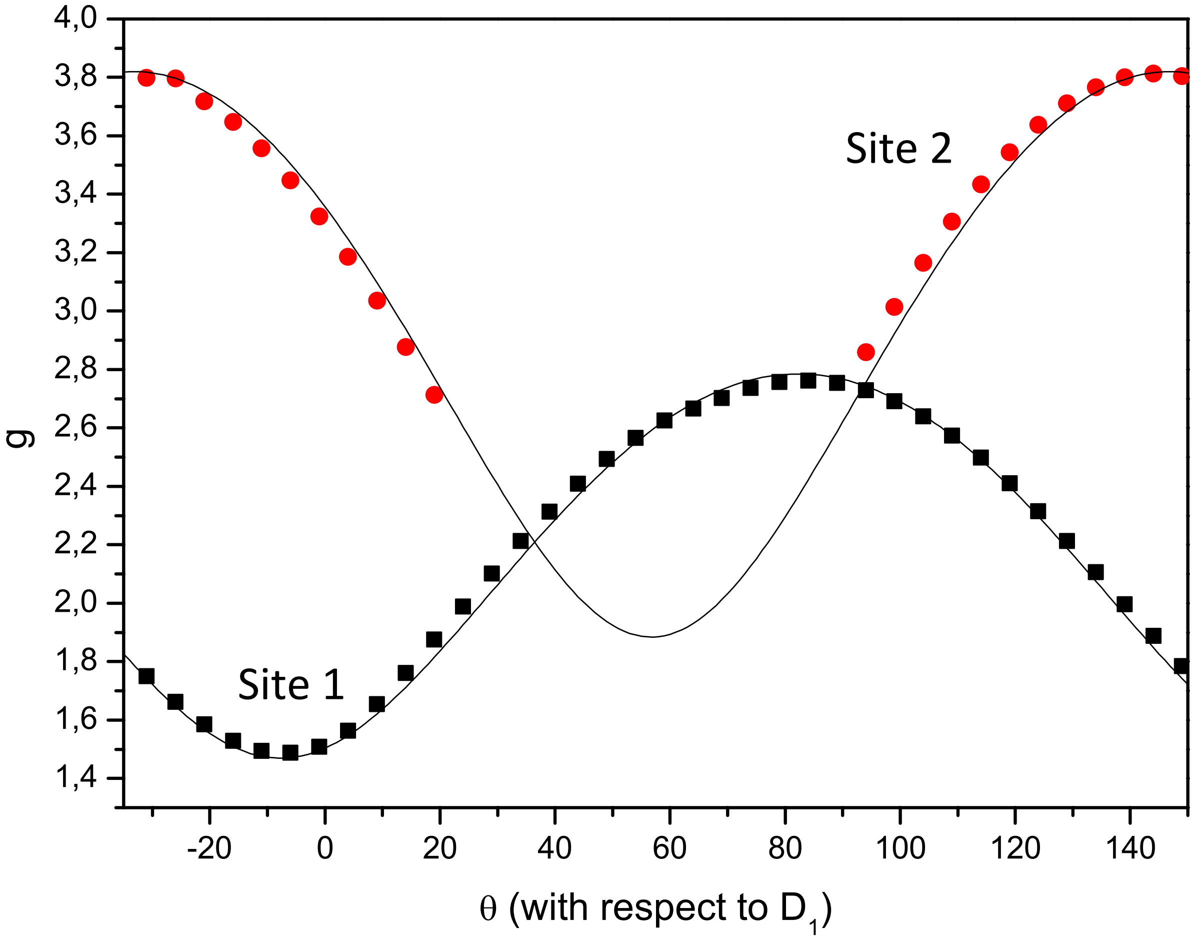
^{143}Nd 12.2%
^{145}Nd 8.3%

Width of ≈ 3 MHz

CW ESR IN THE D_1D_2 PLANE



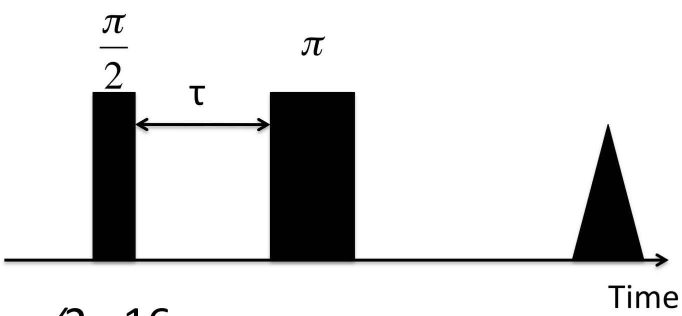
Nd^{3+}	
g_1	g_2
-1.47	0.00(57)
-1.02	1.99
4.20	3.83

Er^{3+} (1)	
g_1	g_2
0.00(5)	0.55(5)
1.79(3)	1.70(3)
14.83	15.54

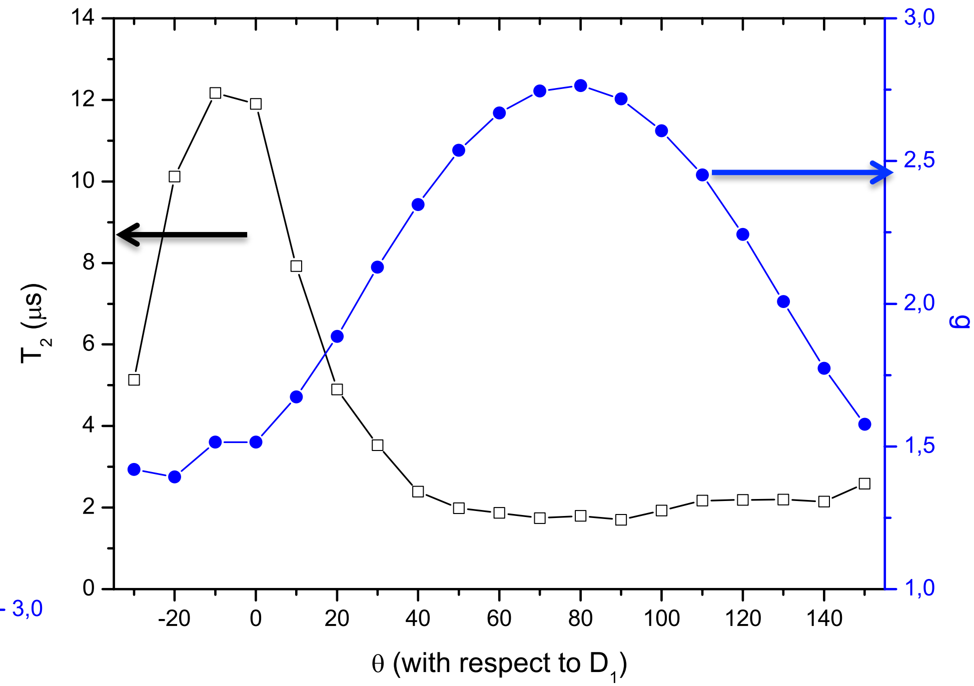
(1) O. Guillot-Noël et al., *PRB*, **74**, 214409, (2006)

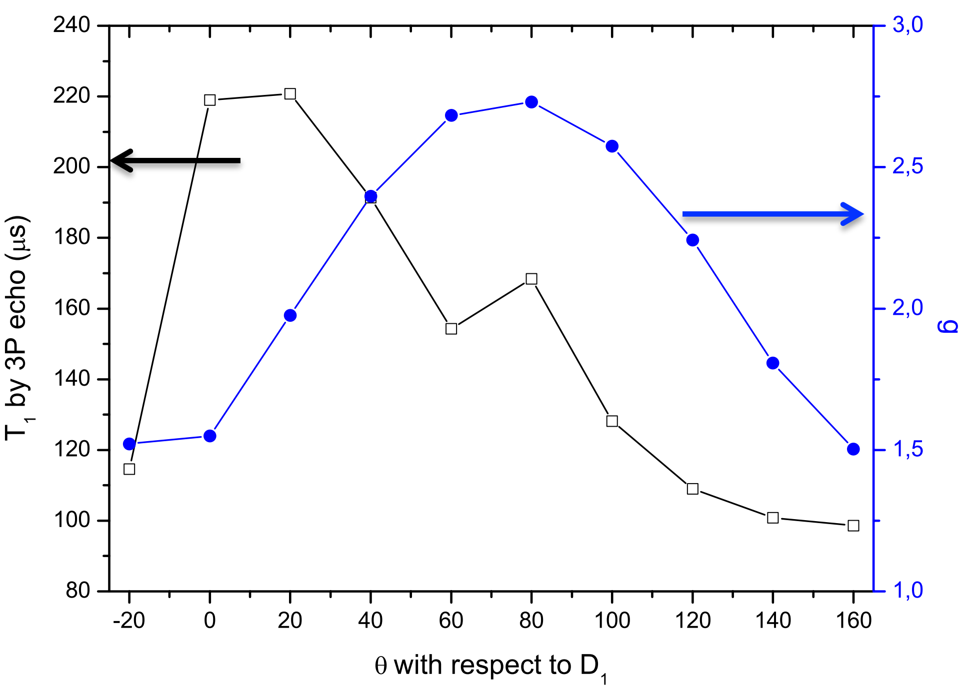
RELAXATION OF ZEEMAN TRANSITIONS

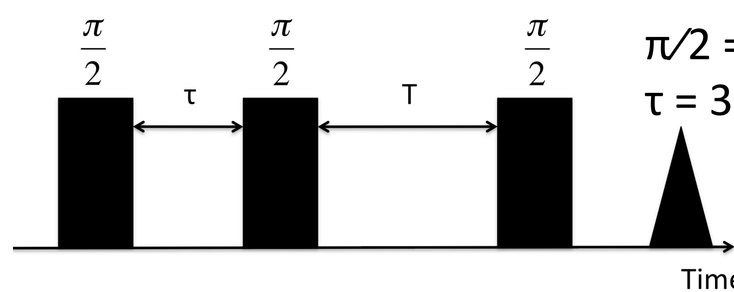
ANISOTROPY OF COHERENCE LIFETIMES



$\pi/2 = 16$ ns



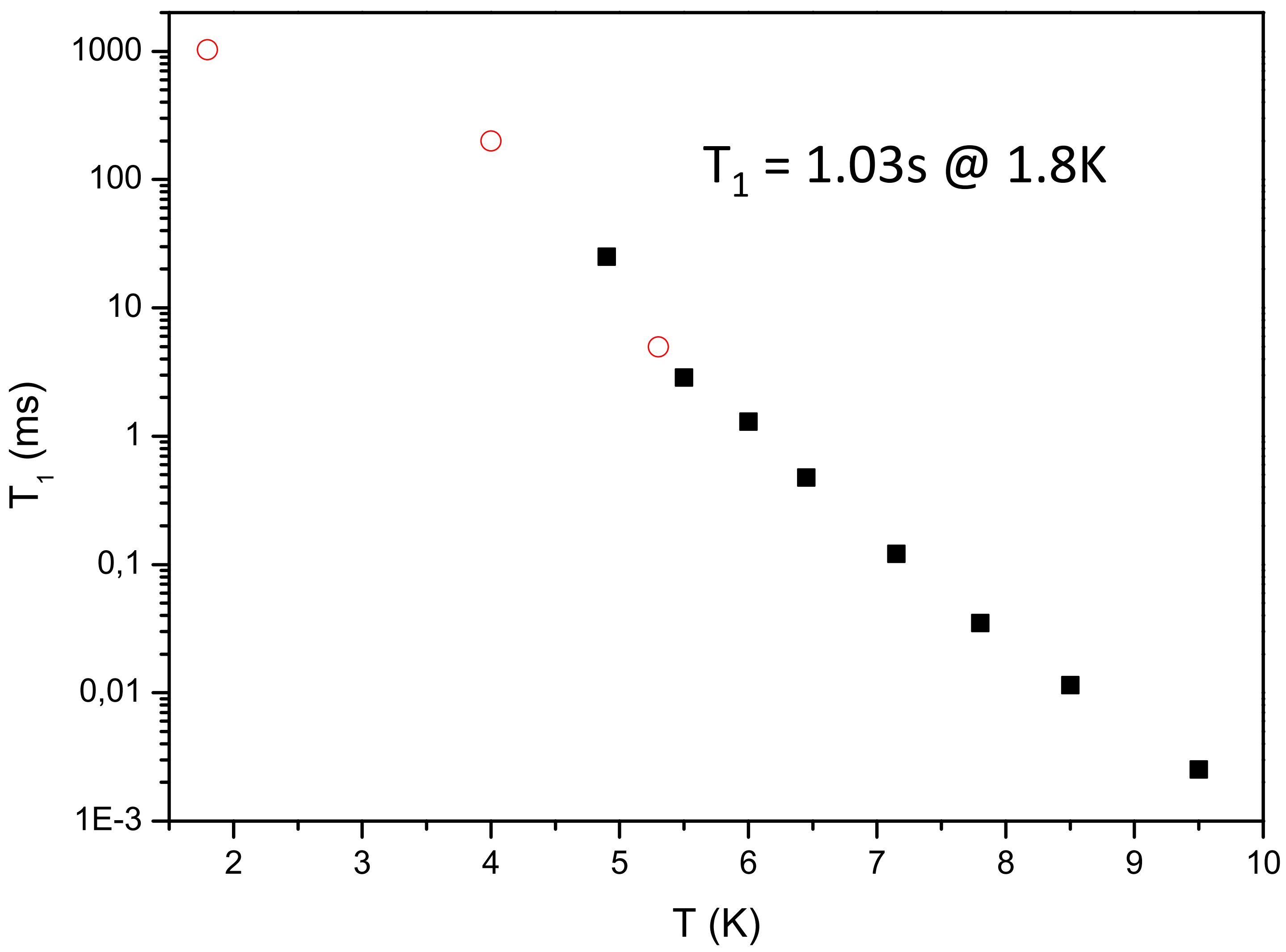




$\pi/2 = 16$ ns
$\tau = 300$ ns

Most efficient orientation : B//D_1

T_1 MEASUREMENT AS A FUNCTION OF THE TEMPERATURE



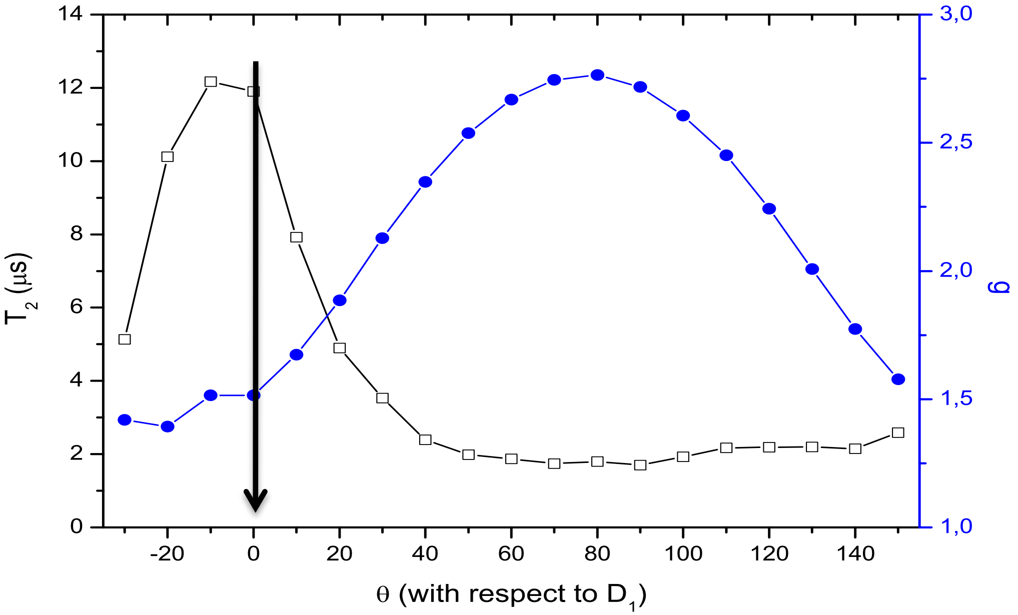
T_2 MEASUREMENT OF HYPERFINE TRANSITIONS

SET UP

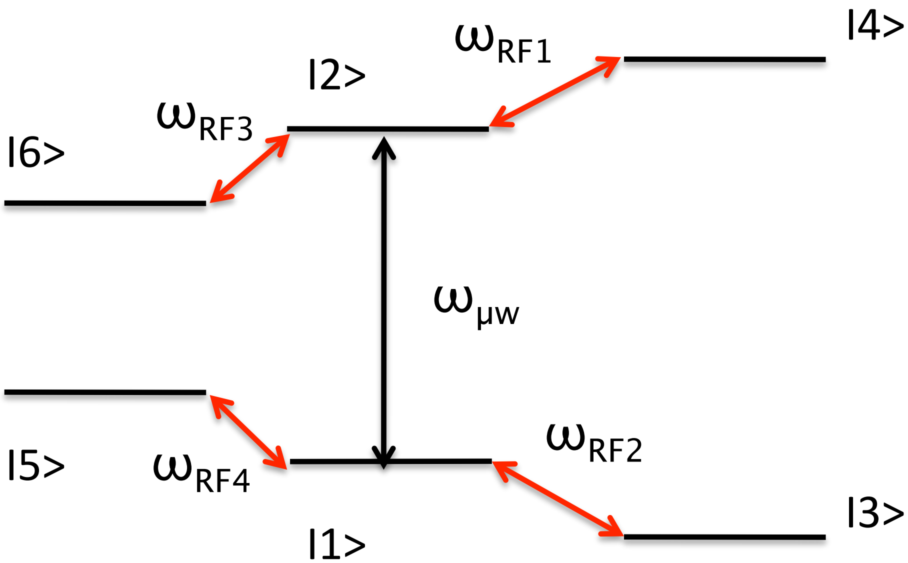
Looking for longest hyp. T_2, B//D1

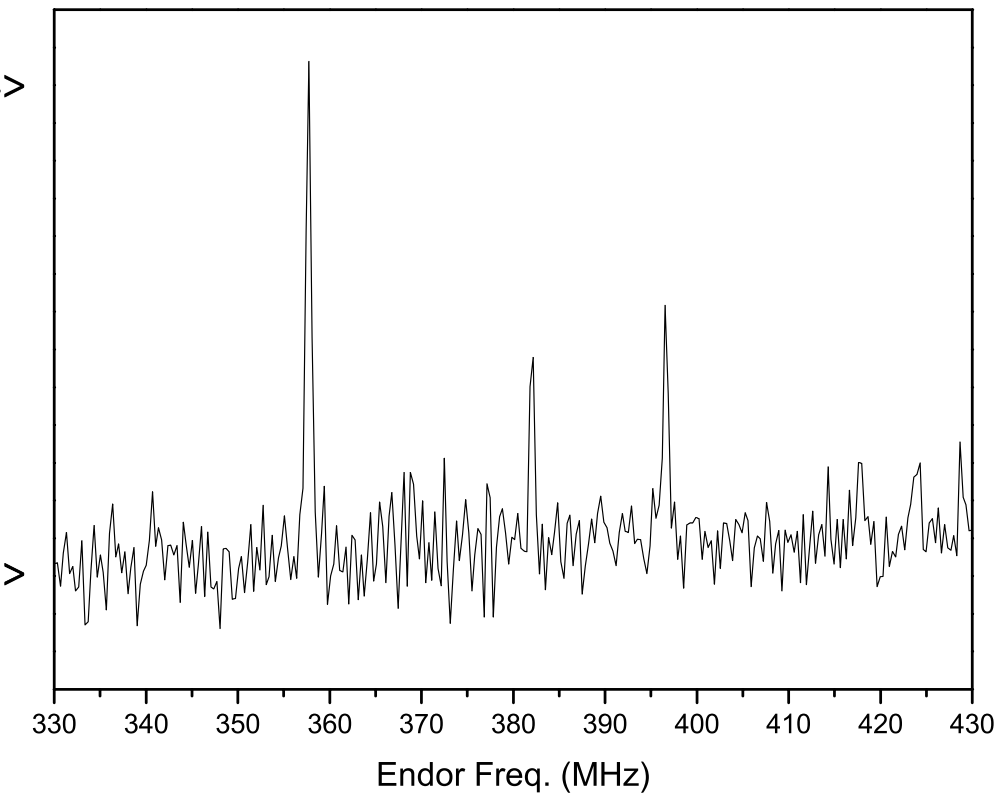
Measured through ENDOR spectroscopy in Oxford (CAESR)

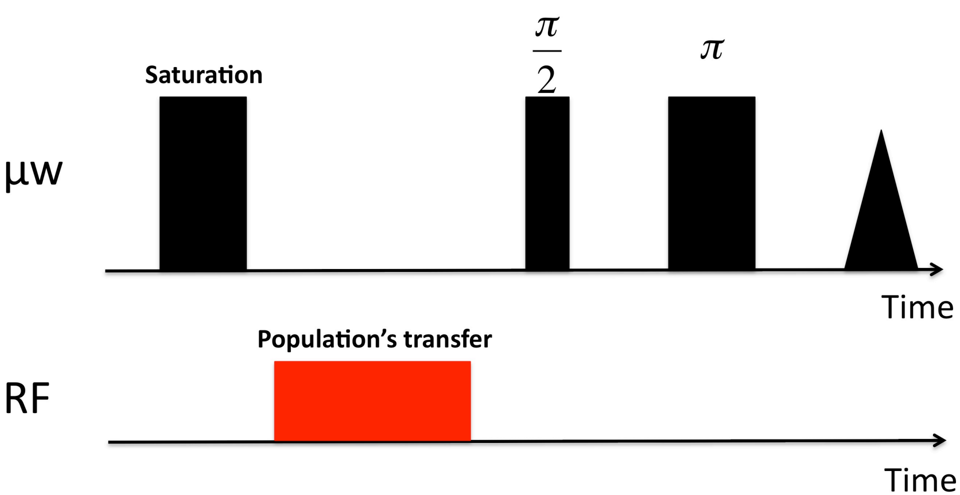
Electron Nuclear DOuble Resonance
Using both μw and RF pulses to address both electronic and nuclear transitions



ENDOR

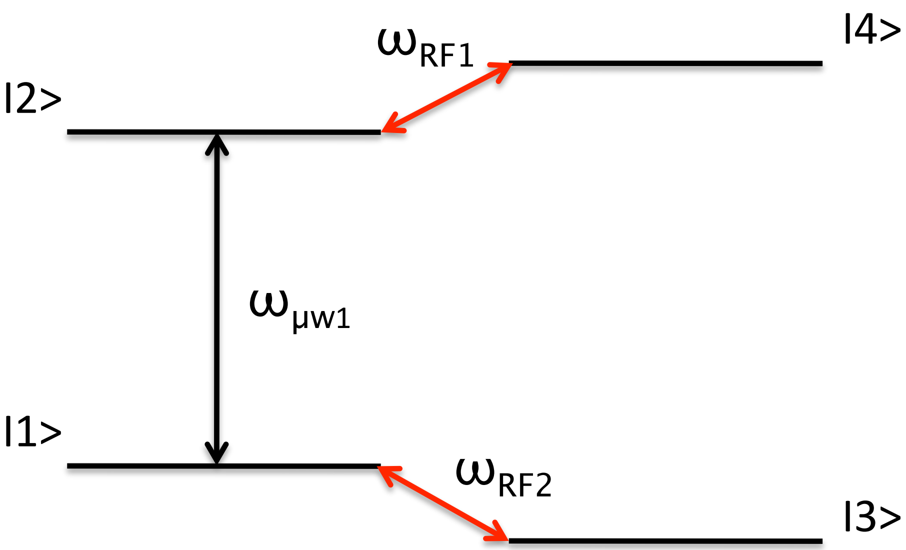


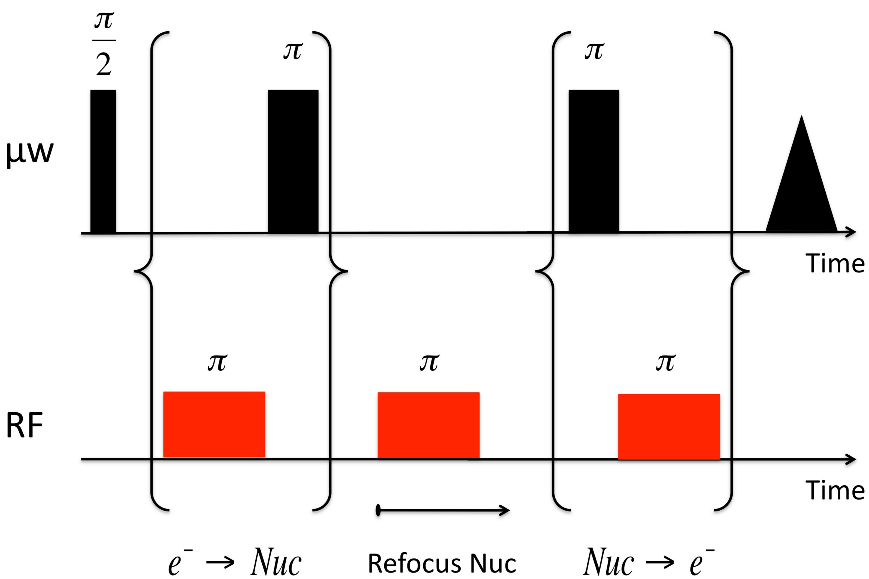


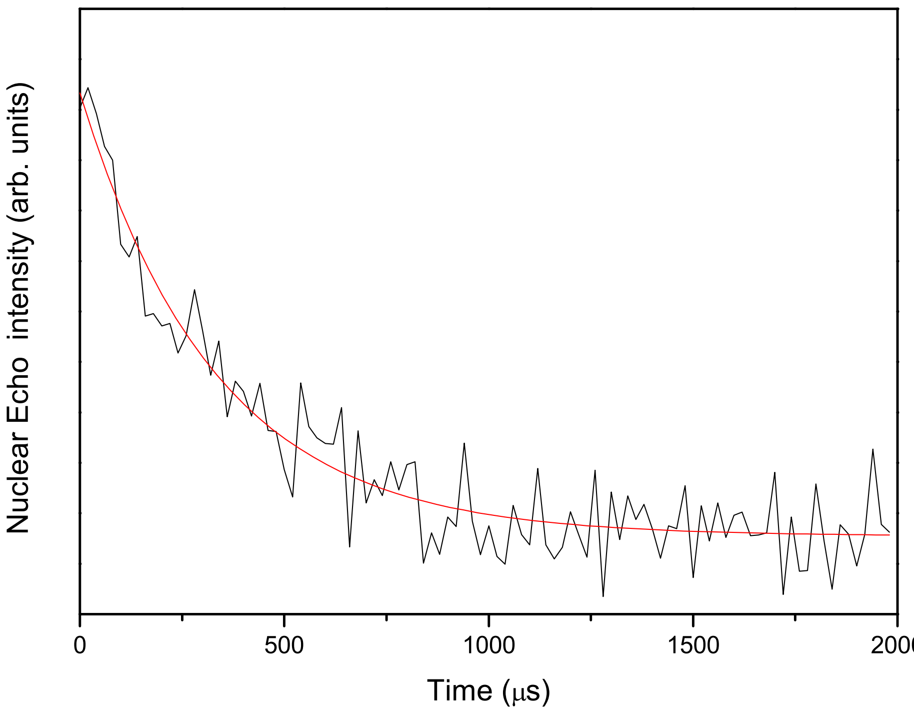


- ○ saturation experiment
- ○ Each Zeeman Line ➔ 4 Endor transitions
- ○ Transitions occur @ 300 – 600 MHz
- ☹ Natural Nd ➔ which isotope?
- ☹ Hyperfine tensor is unknown
- ☹ Not able to reach freq. above 400 MHz

HYPERFINE COHERENCE LIFETIME



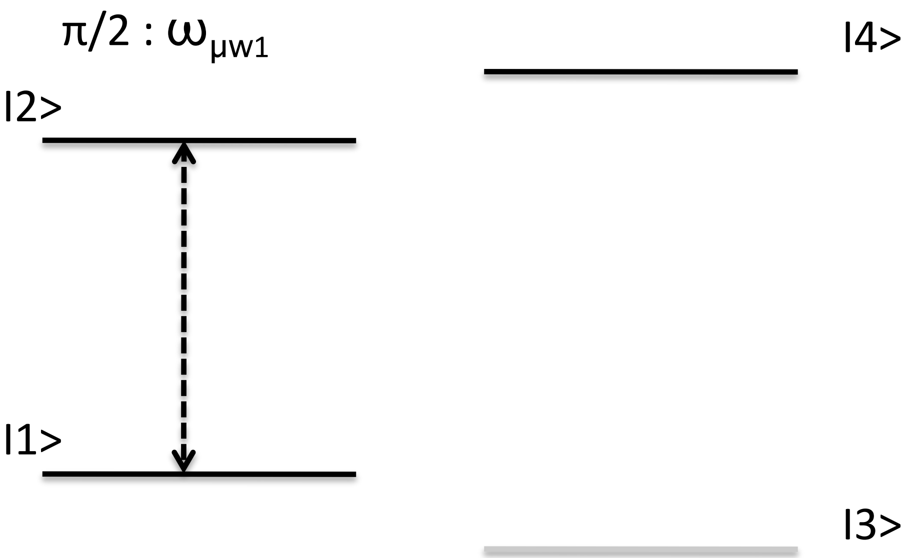


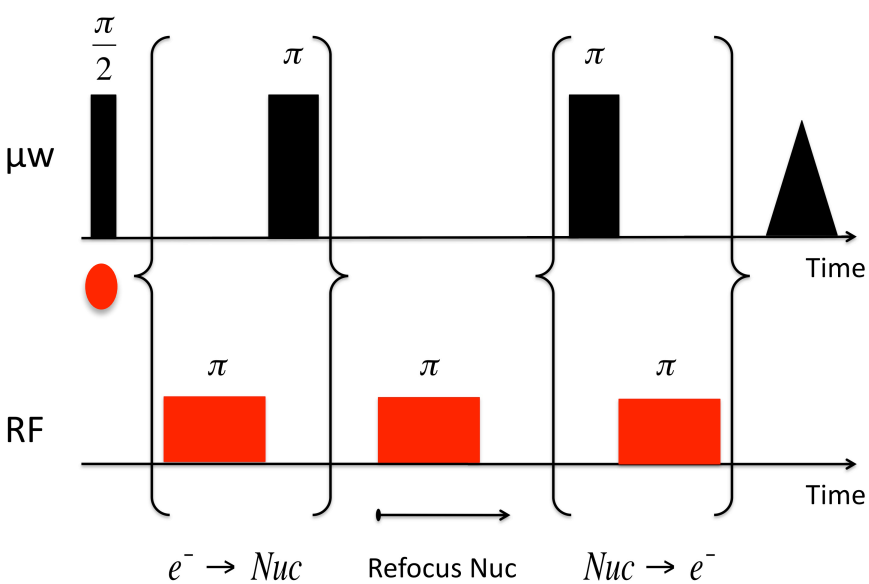


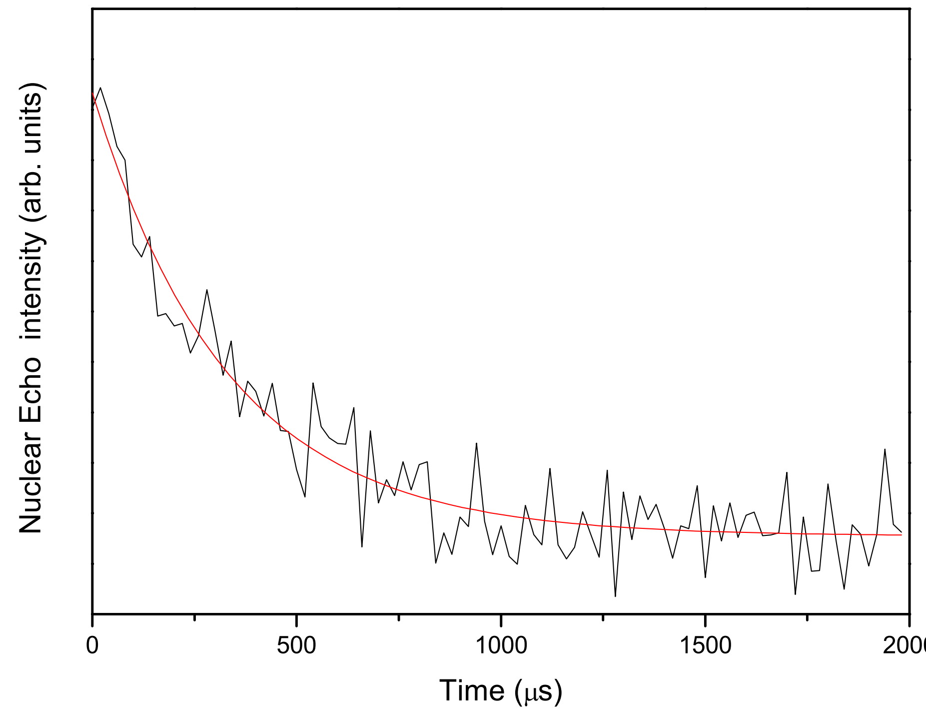
RF Pulses : 3 μs

T_2 = 330 μs at 5.3K

HYPERFINE COHERENCE LIFETIME







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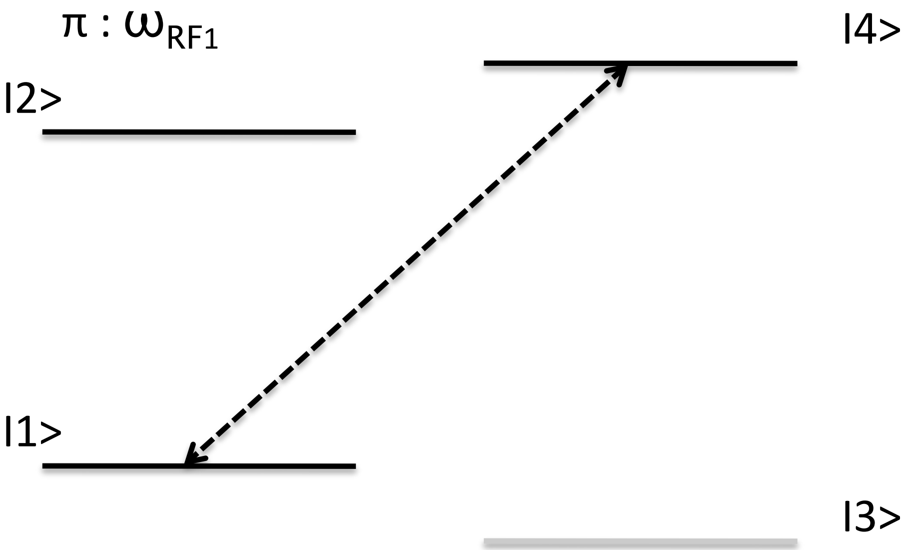

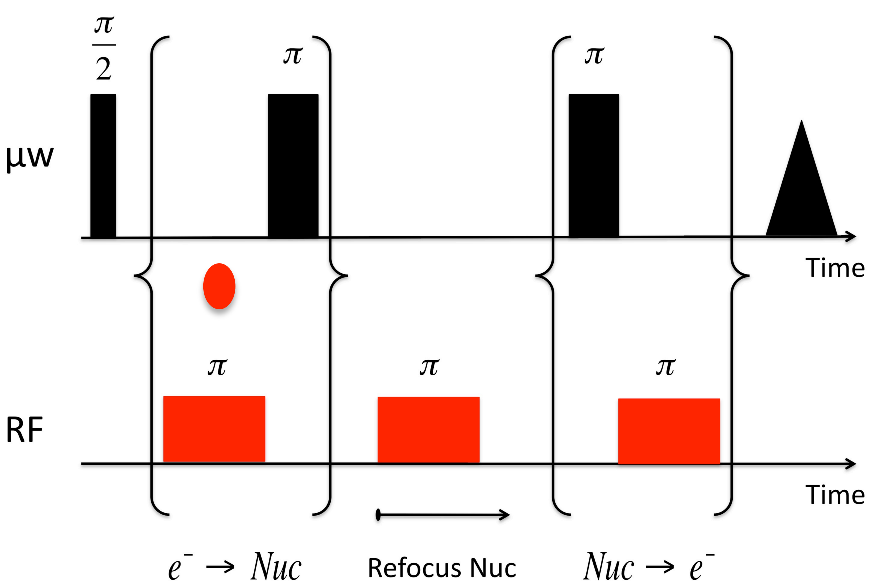

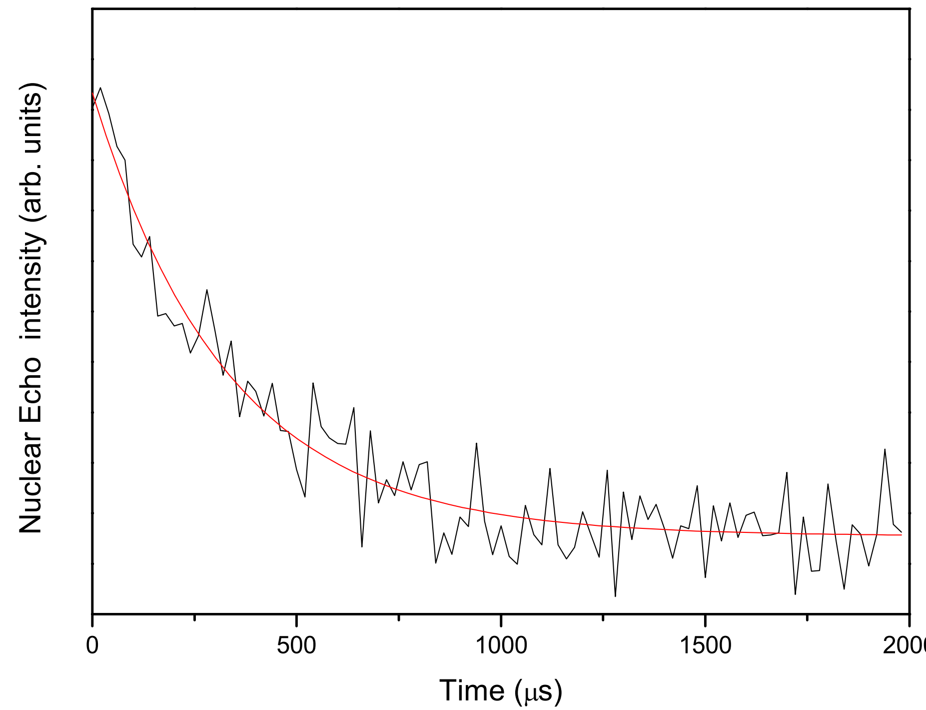

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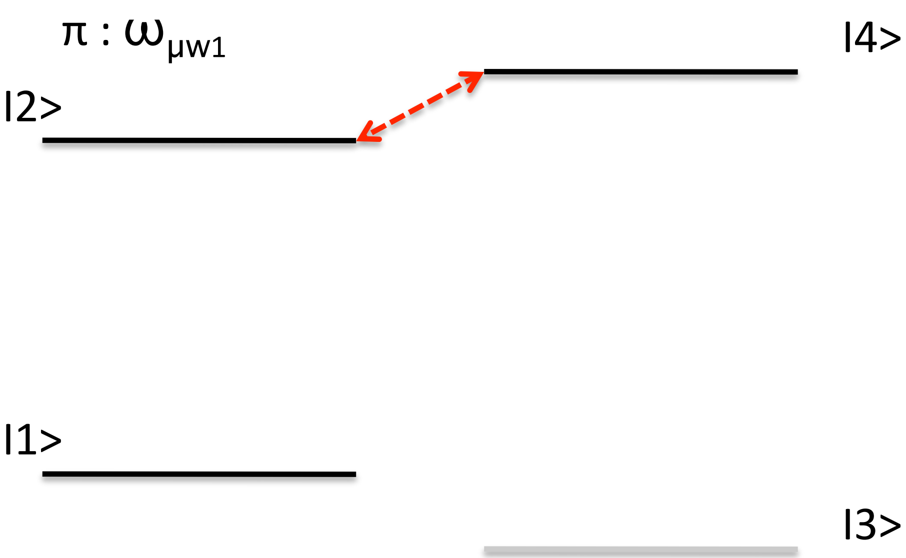

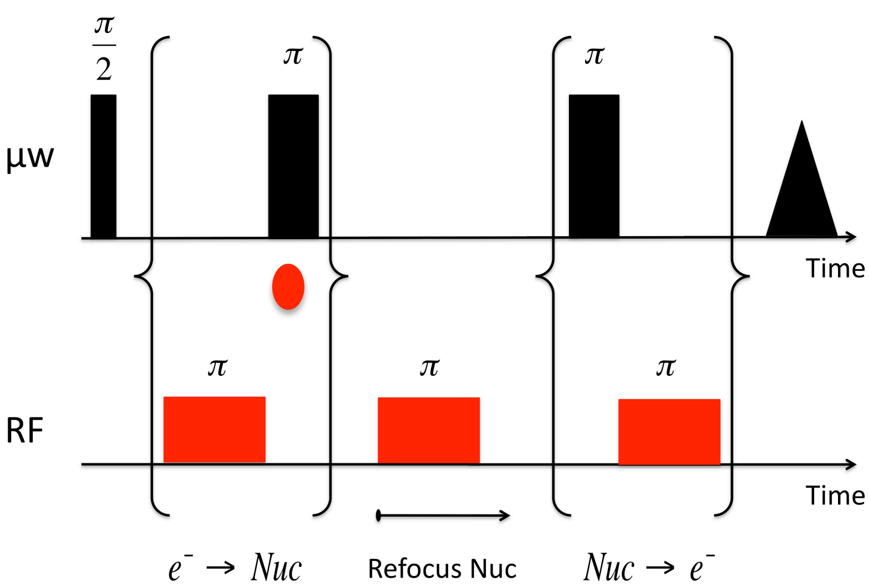

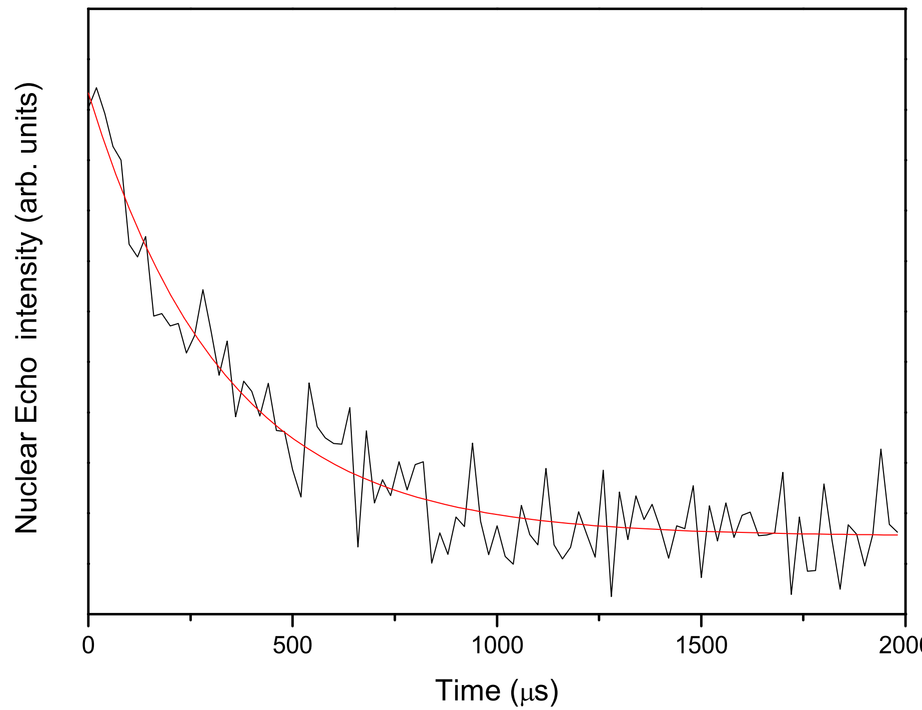

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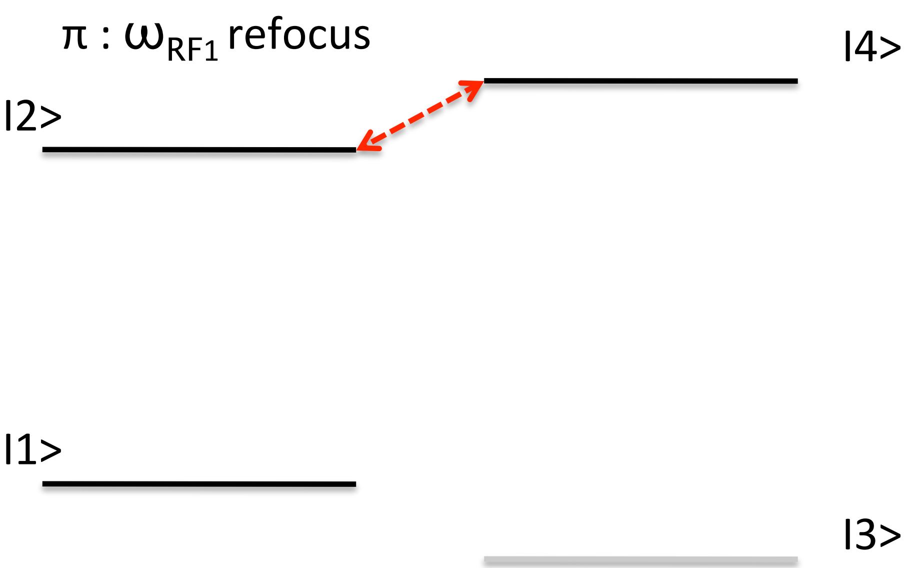

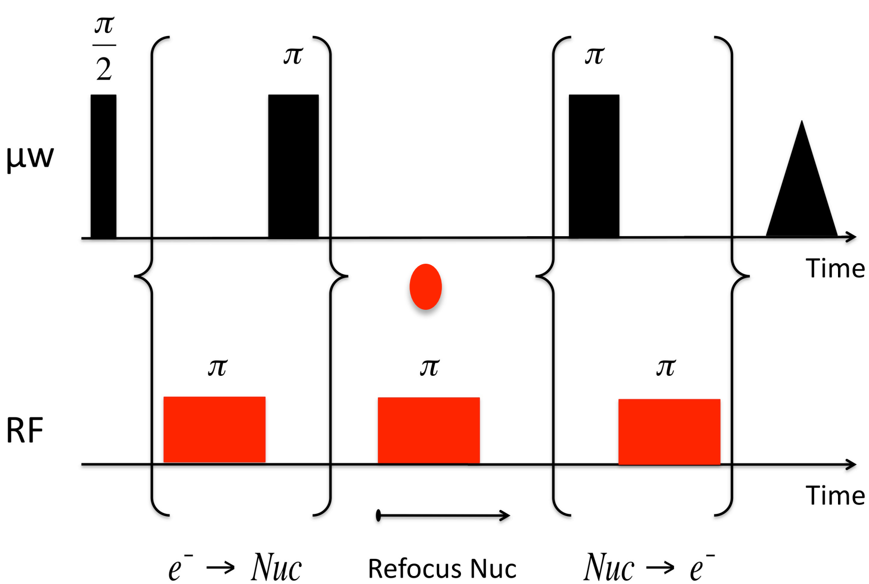

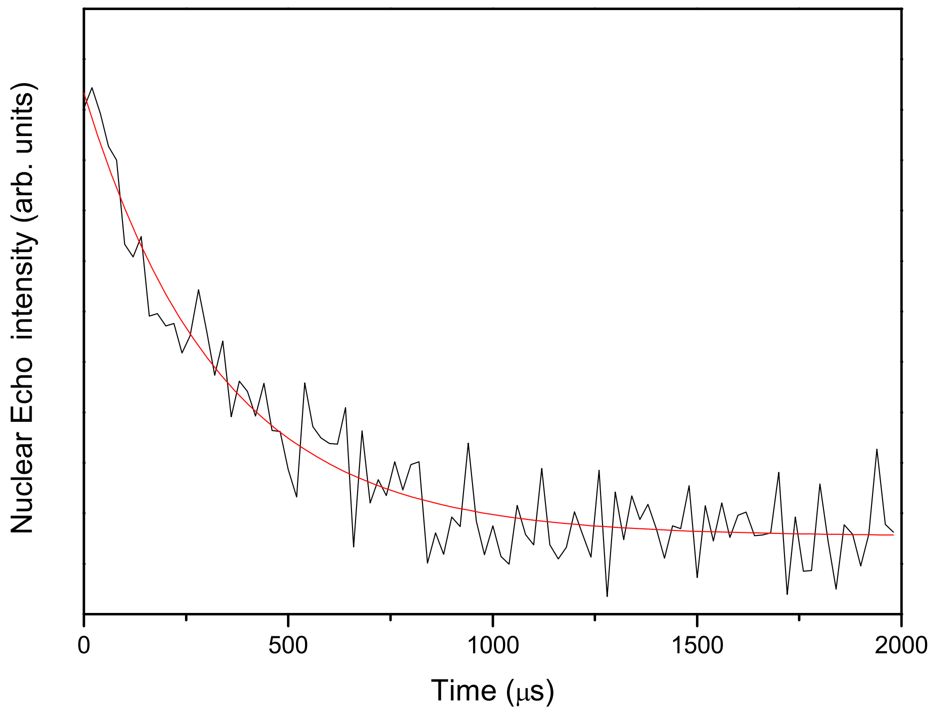

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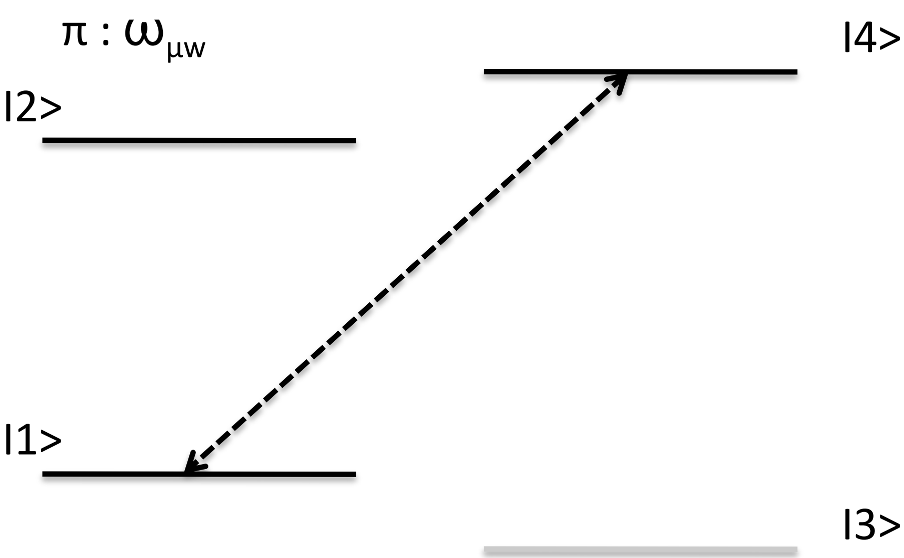

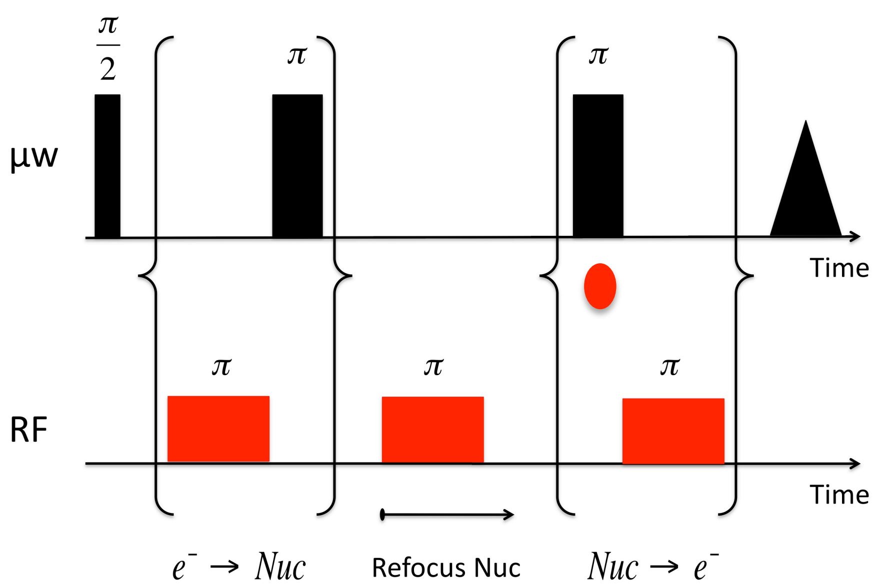

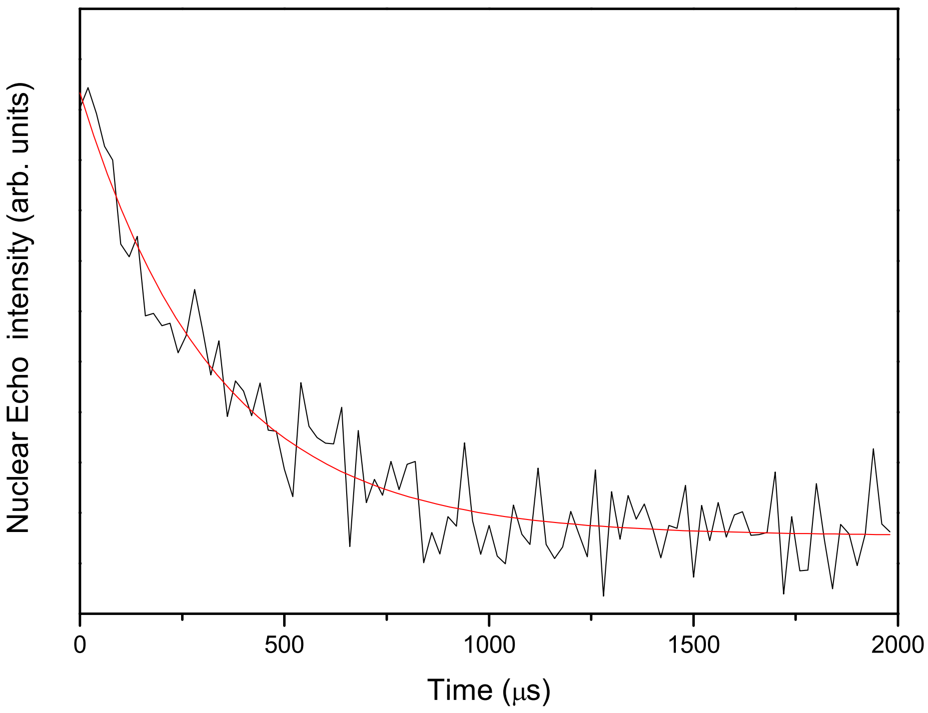

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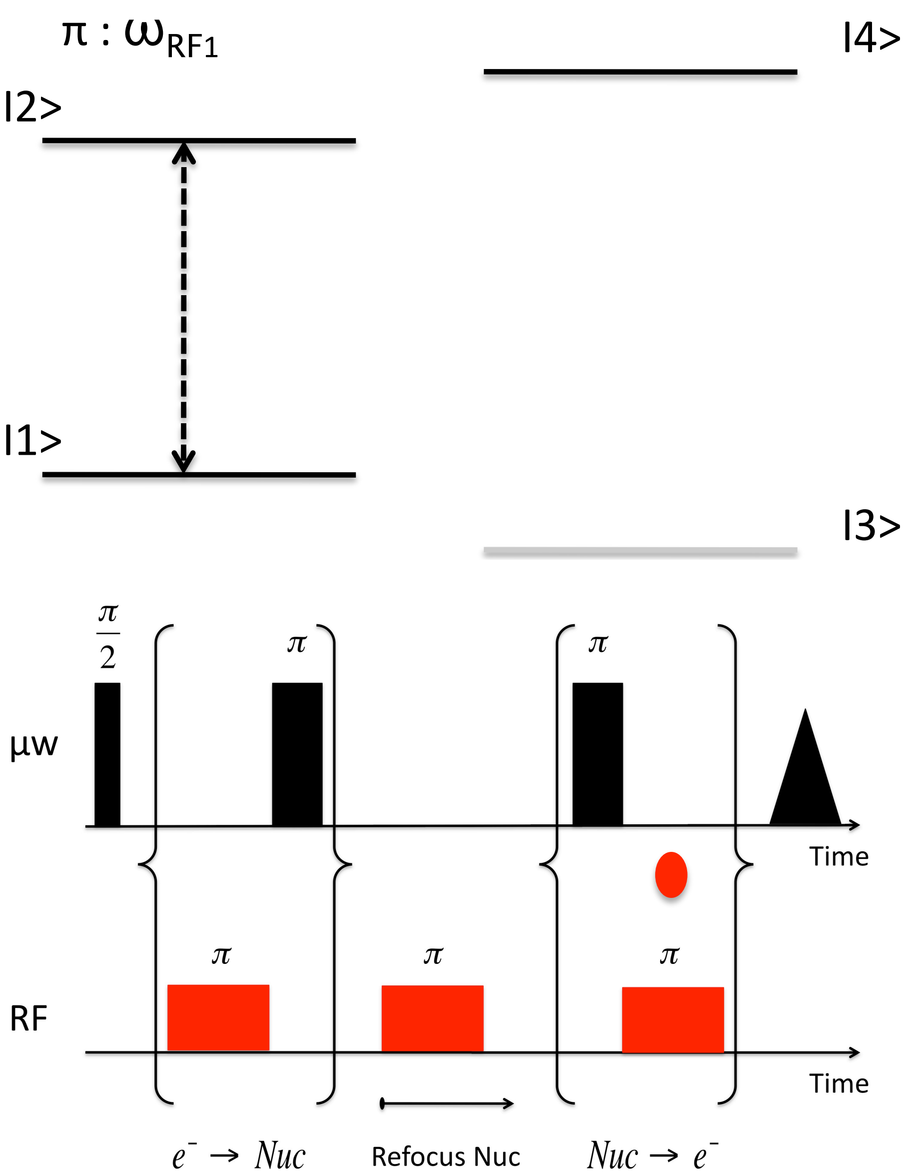
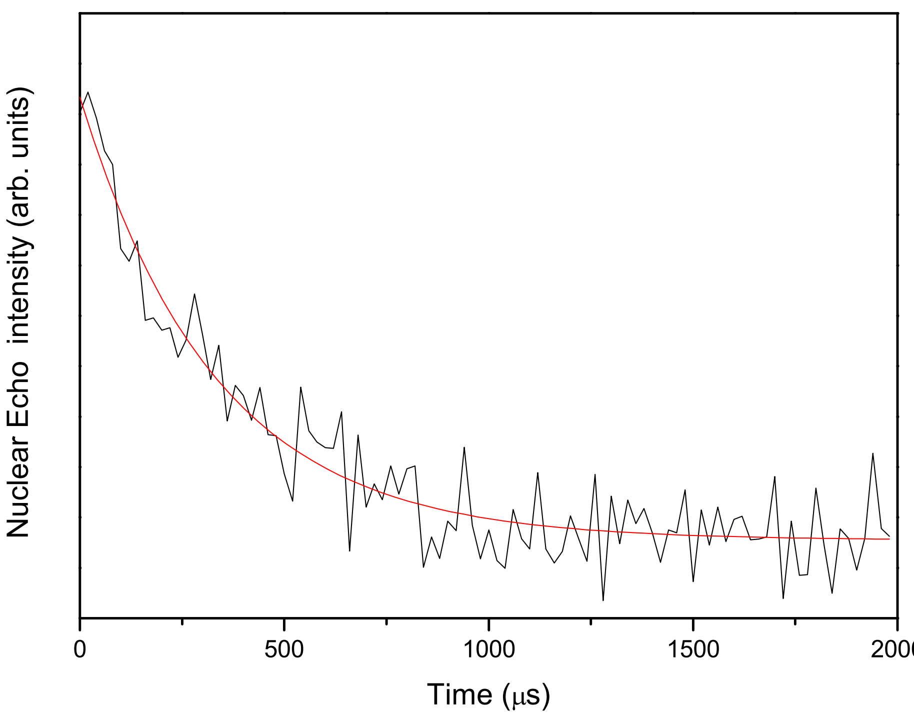




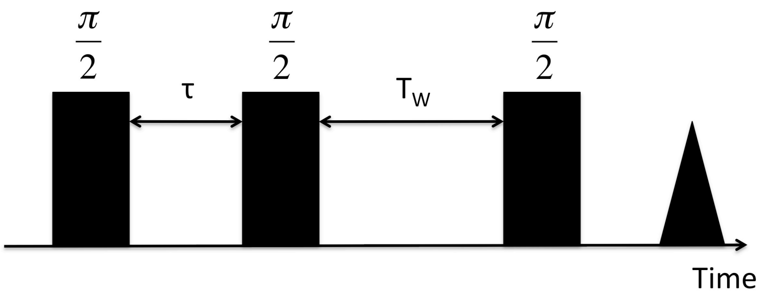
RF Pulses : 3 μs

T_2 = 330 μs at 5.3K

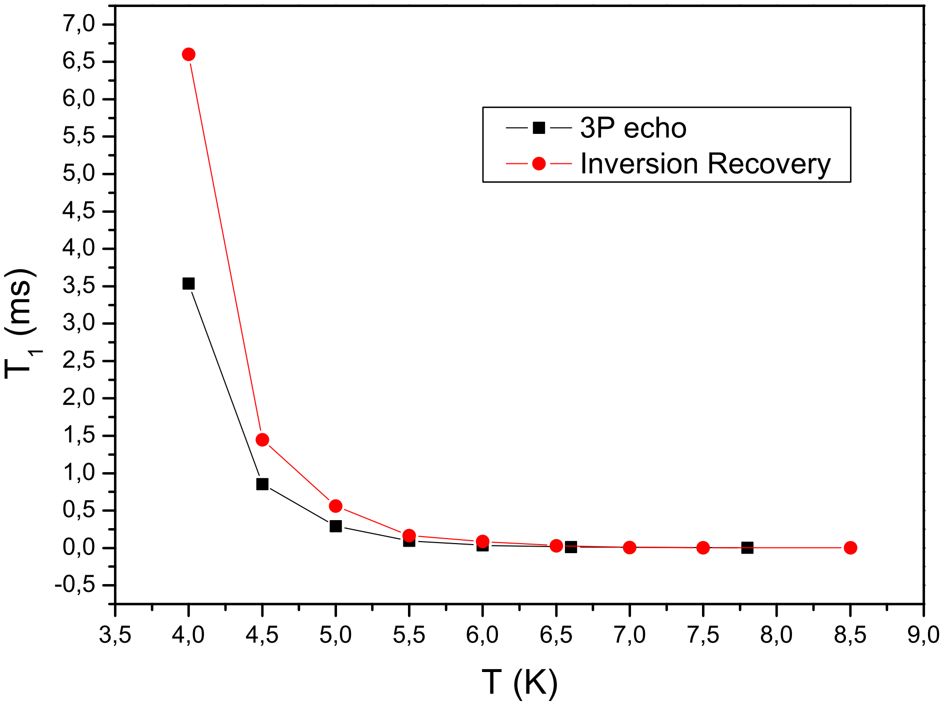
SPECTRAL DIFFUSION

PROOFS OF SPECTRAL DIFFUSION

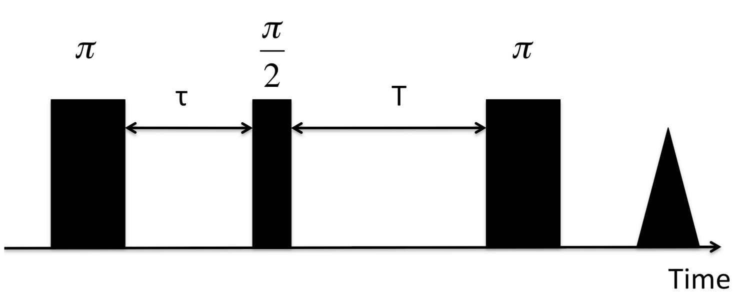
<u>3P Echo</u>



2 first pulses create a spectral grating
The grating evolves during T_W
Sensitive to spectral diffusion



<u>Inversion Recovery</u>



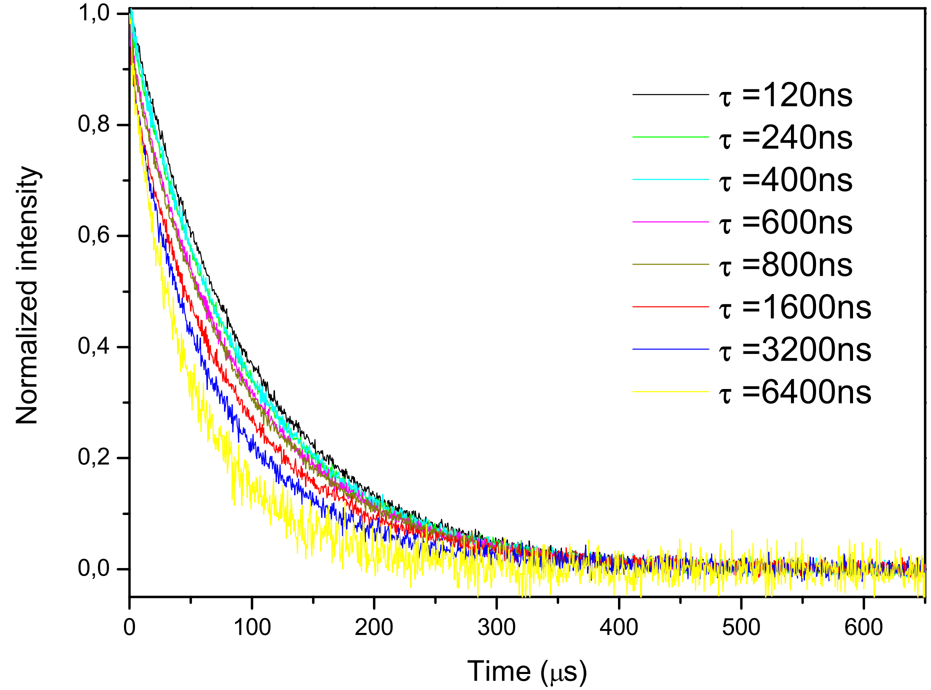
π pulse inverts the population
The population recovers during τ
Pulse is larger than Γ_{inh}
Not-sensitive to spectral diffusion

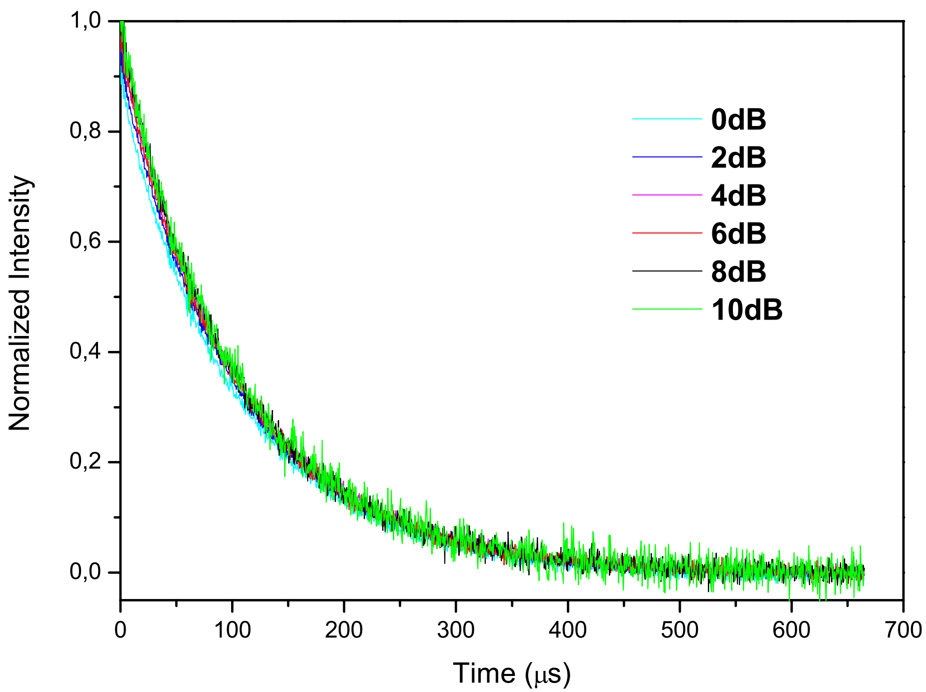
The values of T_1 obtained by the two different methods clearly show evidences of spectral diffusion

PROOFS OF SPECTRAL DIFFUSION

3P Echo measurements
τ varies from 120 to 6400 ns

The shape and lifetime depend on τ
Another evidence of spectral diffusion





3P Echo measurements
Attenuation varies from 0dB to 10dB

The shapes and lifetime do not depend on the microwave power
There is no instantaneous diffusion

TOWARDS A BETTER UNDERSTANDING OF THE SOURCES OF SPECTRAL DIFFUSION

$$A = A_0 \times \exp\left(-\frac{T_W}{T_1}\right) \times \exp\left(-2\pi\Gamma_{eff}\left(T_w,\tau\right)\tau\right)$$

$$\Gamma_{eff}\left(T_w,\tau\right) = \Gamma_0 + \frac{1}{2}\Gamma_{SD}\left[R\tau + \left\{1-\exp\left(-RT_W\right)\right\}\right]$$

T_1: Lifetime of the excited state
Γ_{eff}: Time dependent effective linewidth

Γ_0: Linewidth in absence of SD
Γ_{SD}: FWMH of dynamic distribution of transition frequencies due to dip.-dip. interactions
R: rate of spectral diffusion

Access to :
Major decoherence process (1 or 2 phonon, spin flip, …)
Major source of decoherence

T. Böttger *et al.*, PRB, **73**, 075101, (2006)

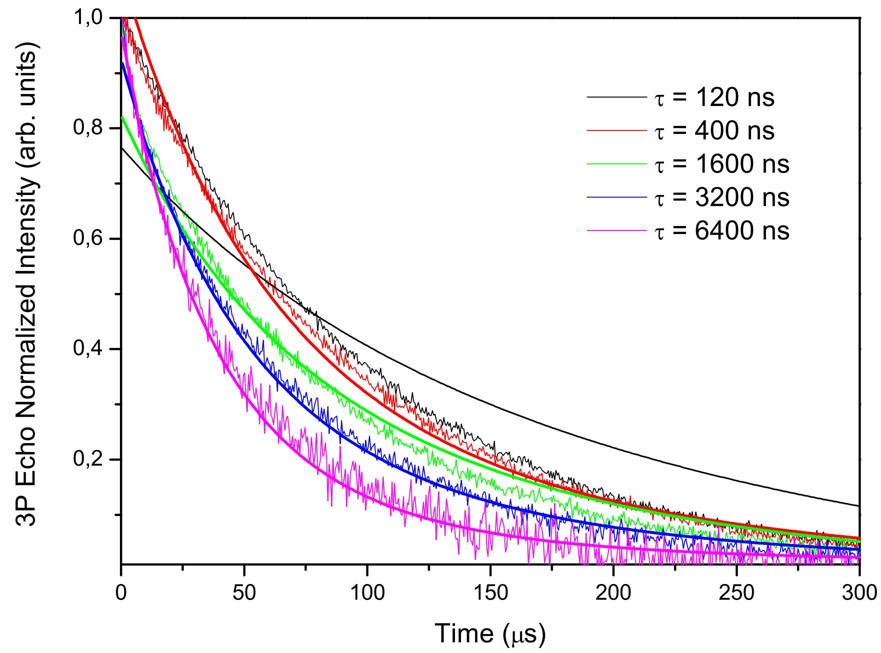
TOWARDS A BETTER UNDERSTANDING OF THE SOURCES OF SPECTRAL DIFFUSION

9 echo decays to determine 3 parameters:

☹ Impossible to find a set of data that allows fitting the 9 decays together

☺ BUT splitting into 2 sub-sets allows finding parameters

☹ Studies are led at high temperature (6.2K)



	Part I	Part II	Exp.
T_1	1.04 ms	1.14 ms	1.27 ms
Γ_0	43 kHz	46 kHz	22.7 kHz
Γ_{SD}	3.09 MHz	3.24 MHz	≈ 3 MHz
R	3.38 kHz	3.99 kHz	1.5 kHz

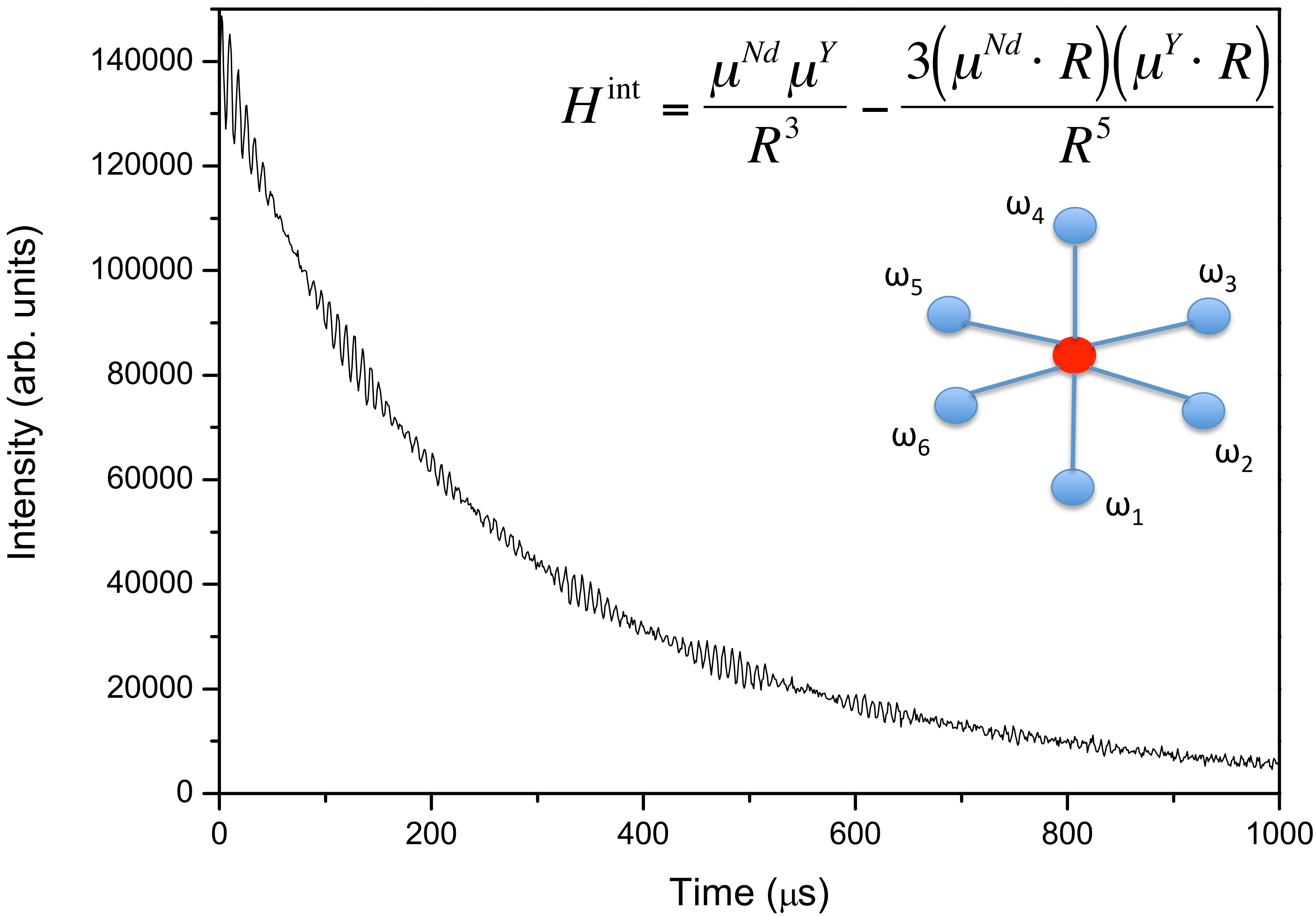
- Nd^{3+} sensitive to the environment
- Other source of SD : nuclear interaction ?

$$\Gamma(t) = \frac{1}{2}\Gamma_N\sqrt{1-\exp(-R_N t)}$$

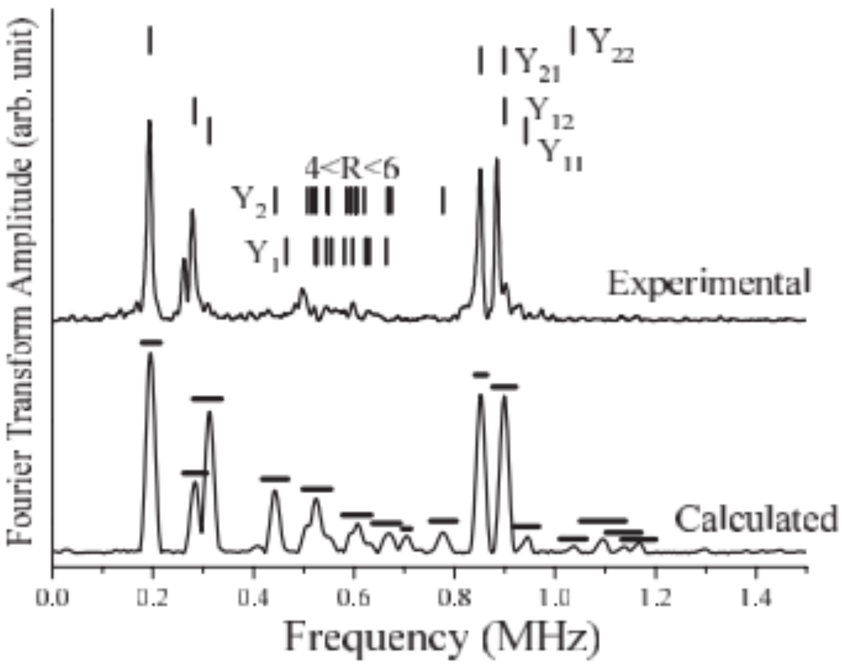
C.W. Thiel et al., *J. Lumin*, **130**, 1603-1609, (2010)

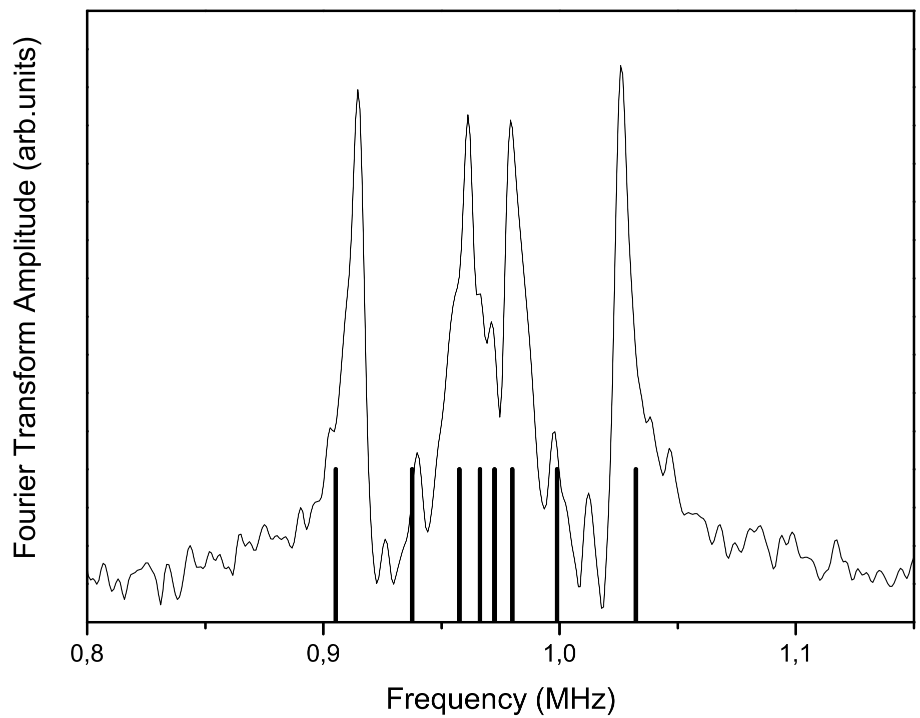
TOWARDS A BETTER UNDERSTANDING OF THE SOURCES OF SPECTRAL DIFFUSION

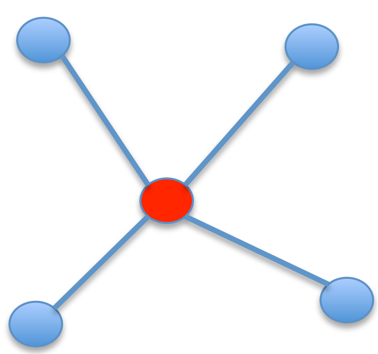
3P ESEEM : Electron Spin Echo Enveloppe Modulation

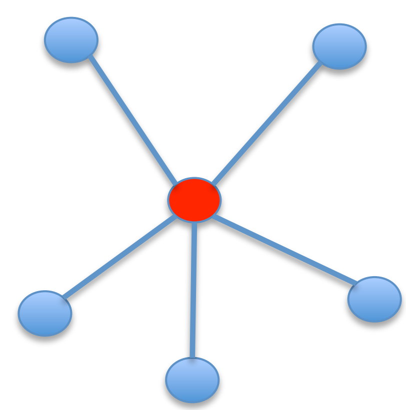


TOWARDS A BETTER UNDERSTANDING OF THE SOURCES OF SPECTRAL DIFFUSION









CONCLUSION

- Main results
  - T_1 = 1.03s @ 1.8K
  - Hyperfine T_2 of 330 μs @ 5K through coherent transfer from electronic to nuclear levels
  - Evidence of spectral diffusion
- About ESR
  - Advantage of ESR: measurement are « easy »
  - Drawback : it is a commercial spectrometer
- Perspectives:
  - Carry on at other magnetic fields
  - Study on an isotopically pure sample (^{145}Nd)
  - Use the results to grow better crystals!

TOWARDS A BETTER UNDERSTANDING OF THE SOURCES OF SPECTRAL DIFFUSION

3P ESEEM : Electron Spin Echo Enveloppe Modulation

$$H = \beta_e B \cdot \tilde{g} \cdot S - \beta_n g_n B \cdot I + H^{\text{int}}$$

$$H^{\text{int}} = \frac{\mu^{Nd}\mu^{Y}}{R^3} - \frac{3\left(\mu^{Nd}\cdot R\right)\left(\mu^{Y}\cdot R\right)}{R^5}$$ Dipole – dipole interaction

$$I(\tau,T) = \frac{1}{2}\left[V^1 + V^2\right]$$ For Nd^{3+} interacting with 1 Y^{3+}

$$V^1 = 1 - \frac{k}{2}\left[1-\cos(\omega_2\tau)\right]\left[1-\cos(\omega_1(\tau+T))\right]$$

$$V^2 = 1 - \frac{k}{2}\left[1-\cos(\omega_1\tau)\right]\left[1-\cos(\omega_2(\tau+T))\right]$$