

### Towards Optically Controlled Qubits in Rare Earth Doped Nanoparticles

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Dept. of Applied Physics and Materials Science, Caltech, May 21, 2018





## Rare Earth Doped Crystals

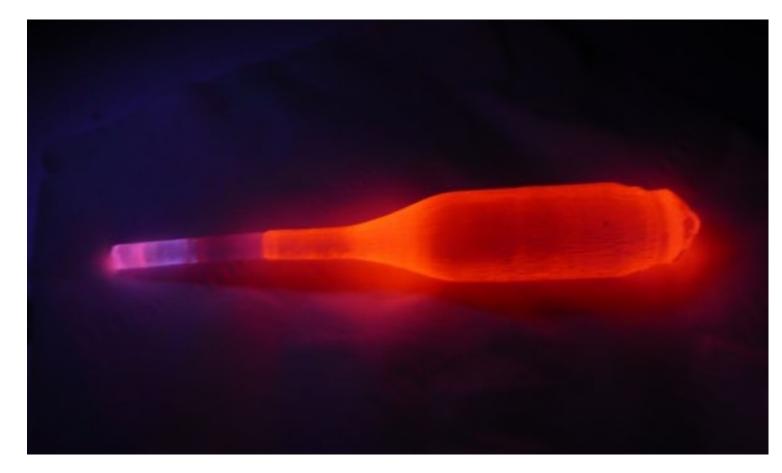
IR

### Lanthanides, scandium, yttrium

1	1/IA 1 H 1.008	2/IIA	~	P	er	0	di	G	[3	b	e		13/IIIA	14/IVA	15/VA	<b>16/</b> VIA		8/VIIIA 2 He 4.003	
2	3 Li 6.941	4 Be 9.012				1998 Dr.	. Michae	al Blabe		5 <b>B</b> 10.81	6 C 12.01	7 N 14.01	8 0 16.00	9 F 19.00	10 Ne 20.18				
3	11 Na 22.99	12 Mg 24.30	◄ VIII → 3/IIIB 4/IVB 5/VB 6/VIB 7/VIIB 8 9 10 11/IB 12/II											14 <b>Si</b> 28.09	15 <b>P</b> 30.97	16 <b>S</b> 32.07	17 Cl 35.45	18 Ar 39.95	
4	19 <b>K</b> 39, 10	20 Ca 40.08	21 SC 44.96	22 <b>Ti</b> 47.87	23 V 50.94	24 Cr 52.00	25 Mn 54.94	26 Fe 55.85	27 Co 58.93	28 Ni 58.69	29 Cu 63.55	30 Zn 65.39	31 Ga 69.72	32 Ge 72.61	33 <b>As</b> 74.92	34 Se 78.96	35 Br 79.90	36 Kr 83.80	
5	37 <b>Rb</b> 85.47	38 <b>Sr</b> 87.62	39 Y 88.91	40 <b>Zr</b> 91.22	41 <b>Nb</b> 92.91	42 <b>Mo</b> 95.94	43 <b>TC</b> 98.91	44 Ru 101.1	45 Rh 102.9	46 Pd 106.4	47 <b>Ag</b> 107.9	48 Cd 112.4	49 In 114.8	50 <b>Sn</b> 118.7	51 Sb 121.8	52 <b>Te</b> 127.6	53   126.9	54 Xe 131.3	
6	55 Cs 123.9	56 Ba 137.3	La- Lu	72 Hf 178,5	73 <b>Ta</b> 180.9	74 W 183.8	75 <b>Re</b> 186.2	76 <b>OS</b> 190.2	77 <b>Ir</b> 192.2	78 Pt 195.1	79 Au 197.0	80 Hg 200.6	81 <b>TI</b> 204.4	82 Pb 207.2	83 <b>Bi</b> 209.0	84 <b>Po</b> 210.0	85 At 210.0	86 <b>Rn</b> 222.0	
7	87 <b>Fr</b> 223.0	88 Ra 226.0	Ac- Lr	104 Db	105 JI	106 Rf	<sup>107</sup> Bh	108 Hn	109 Mt	110 Uun	111 Uuu								
	<b>←</b> s		•	<i>d</i>							<b>*</b> 4				p				
Lanthanides Actinides				57 La 138.9	58 Ce 140.1	59 <b>Pr</b> 140.9	60 Nd 144.2	61 Pm 146.9	62 Sm 150.4	63 Eu 152.0	64 Gd 157.2	65 <b>Tb</b> 158.9	66 <b>Dy</b> 162.5	67 <b>Ho</b> 164.9	68 Er 167.3	69 <b>Tm</b> 168.9	70 Yb 173.0	71 Lu 175.0	
				89 Ac 227.0	90 Th 232.0	91 Pa 231.0	92 U 238.0	93 Np 237.0	94 Pu 239.1	95 Am 241.1	96 Cm 244.1	97 Bk 249.1	98 Cf 252.1	99 Es 252.1	100 Fm 257.1	101 Md 258.1	NO 259.1	103 Lr 262.1	
				•								<i>f</i> — •							

#### Stable centres, no bleaching

### Single crystals, films, particles

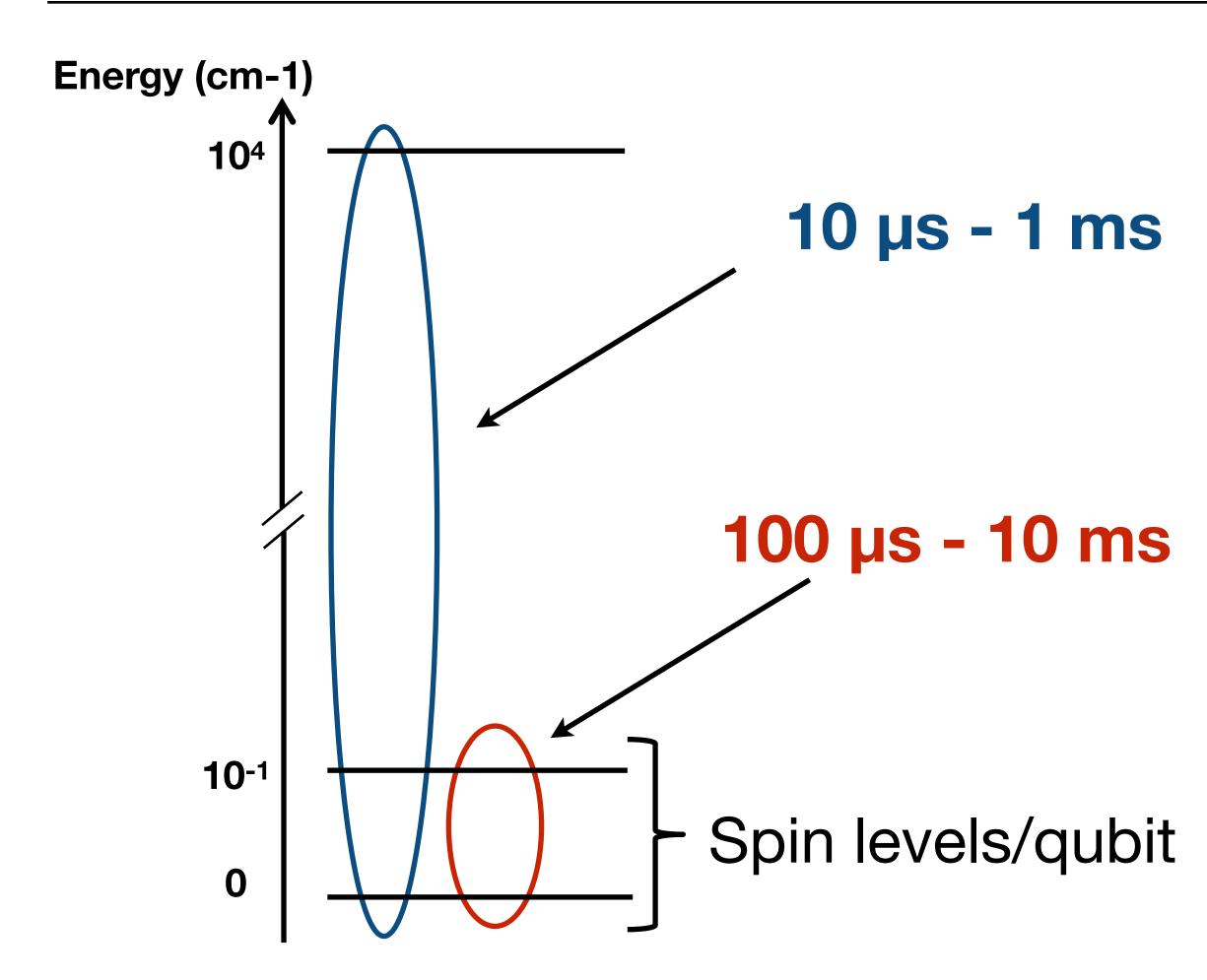


Y<sub>2</sub>SiO<sub>5</sub>:Eu<sup>3+</sup> emission under UV excitation

Lasers, phosphors, bio-probes...

## **Energy Levels and Transitions**

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P. Goldner, A. Ferrier, and O. Guillot-Noël, in Handbook on the Physics and Chemistry of Rare Earths, vol. 46, 2015 Optical transitions in the visible and infrared range

Screening of 4f electrons: long optical T<sub>2</sub> (at LHe temp)

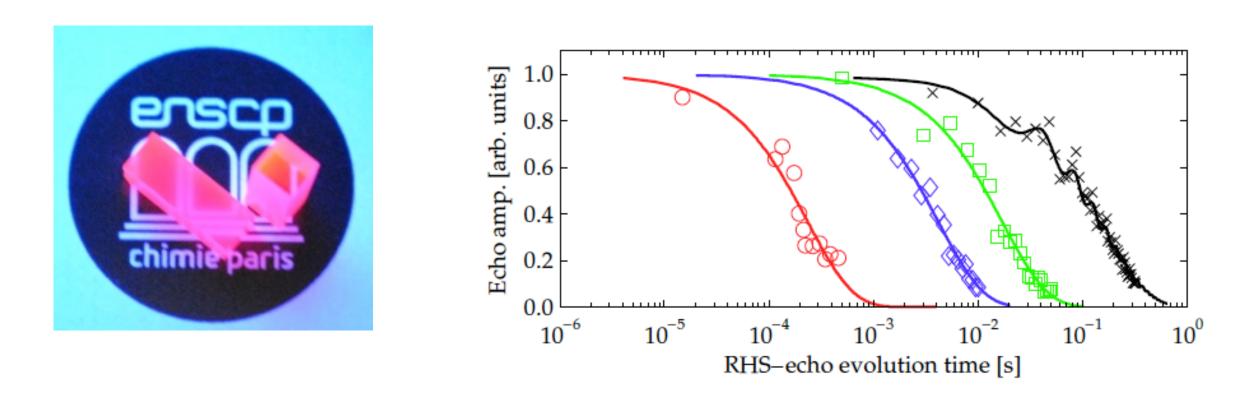
Electron and/or nuclear spins

Optical quantum memories in bulk crystals and fibres

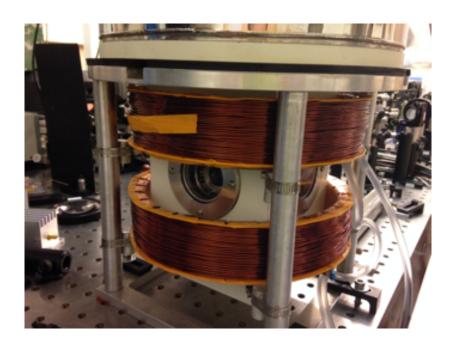
W. Tittel et al., Nature Photon., 2009. M. Zhong et al., Nature, 2015.

### **Crystals and Quantum State Dynamics**

### Designing and growing crystals with long lived quantum states **Controlling quantum states dynamics** by external fields



#### Crystal growth **bulk**, **particles**, **thin films High resolution and** coherent spectroscopy



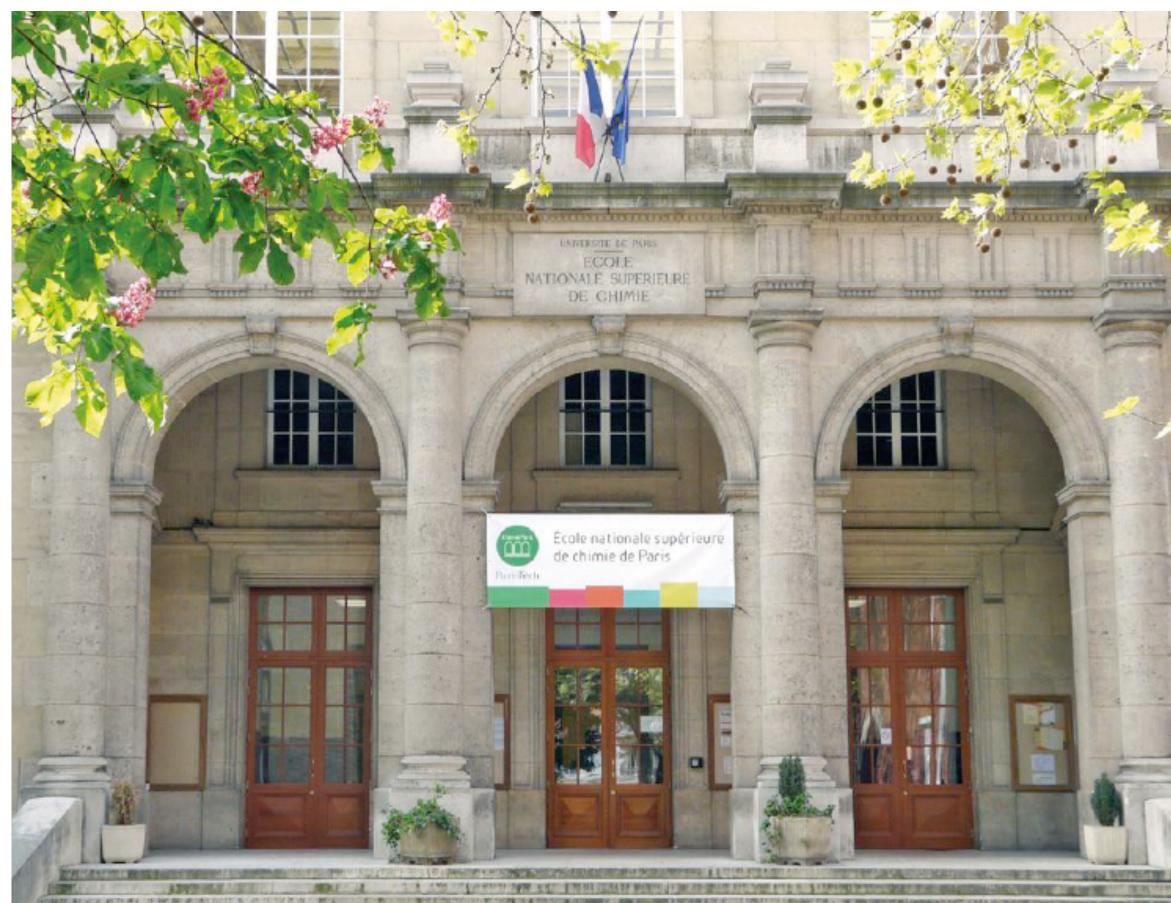
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**Quantum information processing** High bandwidth signal processing Ultrasound optical tomography Ultra stable laser locking







### Chimie ParisTech



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Chimie Paristech founded in 1896 moved to the present building in the Latin quarter in 1920

About 300 students follow lectures in all fields of chemistry

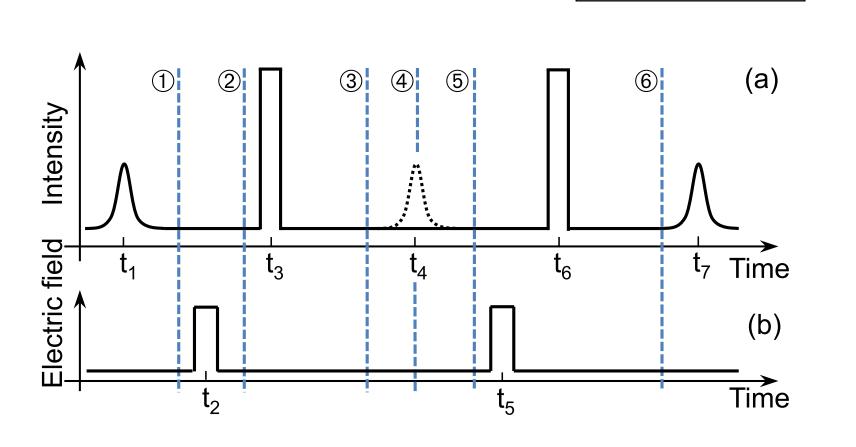
**Paris Institute for Chemical Research:** about 200 researchers and PhD students

## **Recent Results with RE Spins**

### Electric field effects

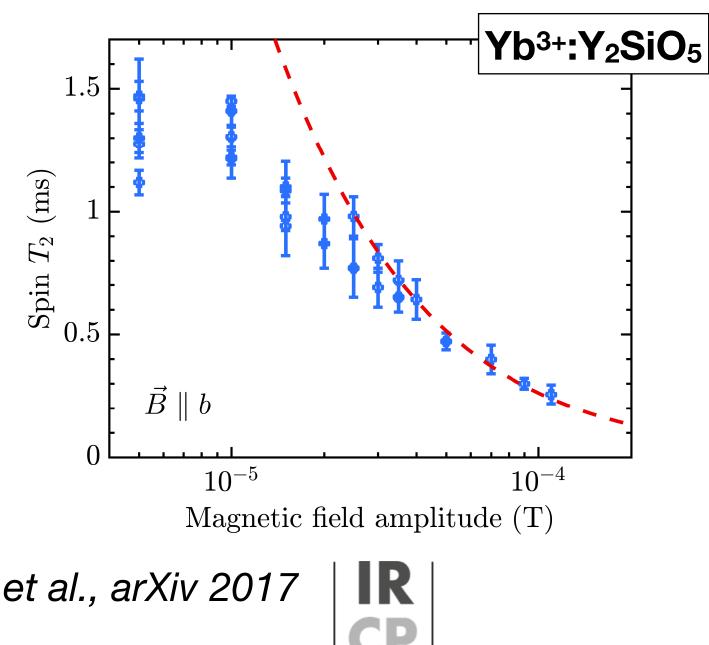
T<sub>2</sub> extension

### Stark shift on RE spins Qu. memory protocol



**Eu<sup>3+</sup>:Y<sub>2</sub>SiO**<sub>5</sub>

### Trains of RF pulses **Clock transitions**

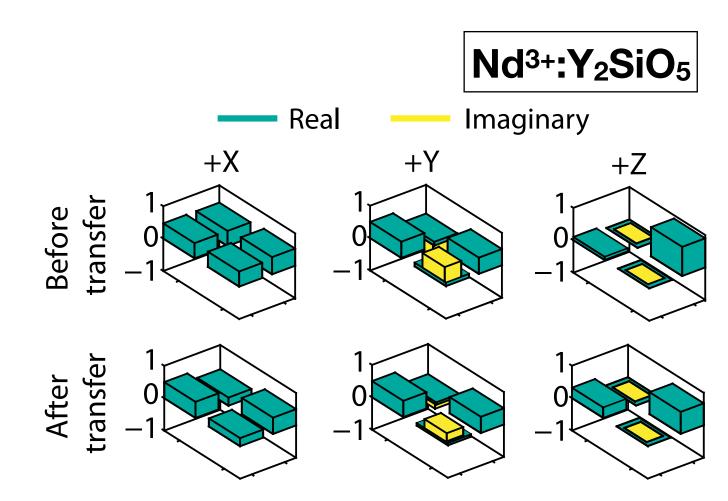


A. Arcangeli et al., PRA 2016 R.M. Macfarlane et al. PRL 2014.

A. Tiranov et al., arXiv 2017

### Storage

### Optical and **microwave** excitations to spin



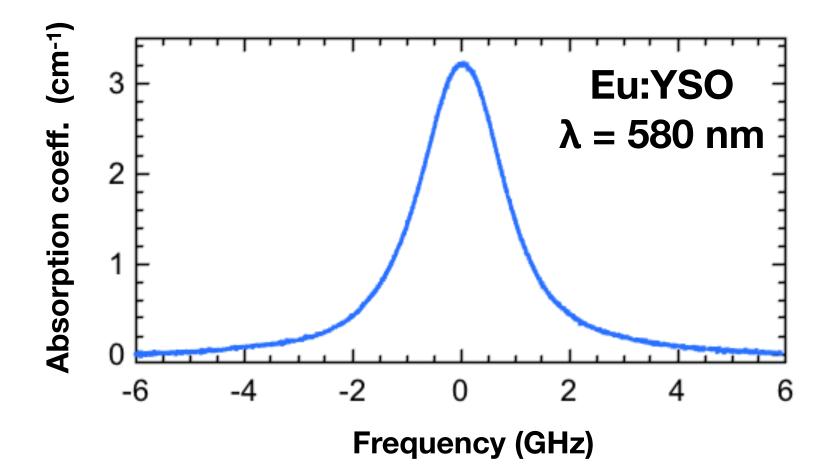
G. Wolfowicz et al., PRL 2015. M. Lovrić et al., PRL 2013.



### Rare Earth Ions as Qubits

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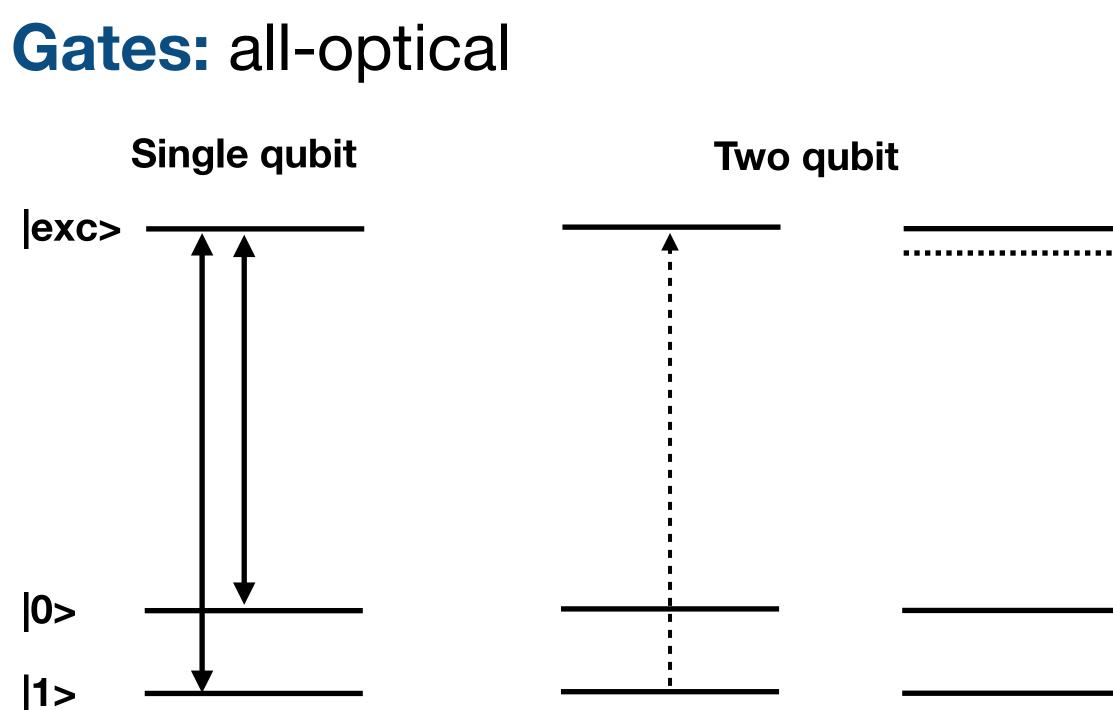
### Scalability: optical addressing



Ensemble linewidth  $\Gamma_{inh} \approx 2 \text{ GHz}$ Single ion linewidth  $\Gamma_h \approx 1 \text{kHz}$ 

### 10<sup>4</sup> qubits!!

N. Ohlsson et al., Opt. Commun., 2002.



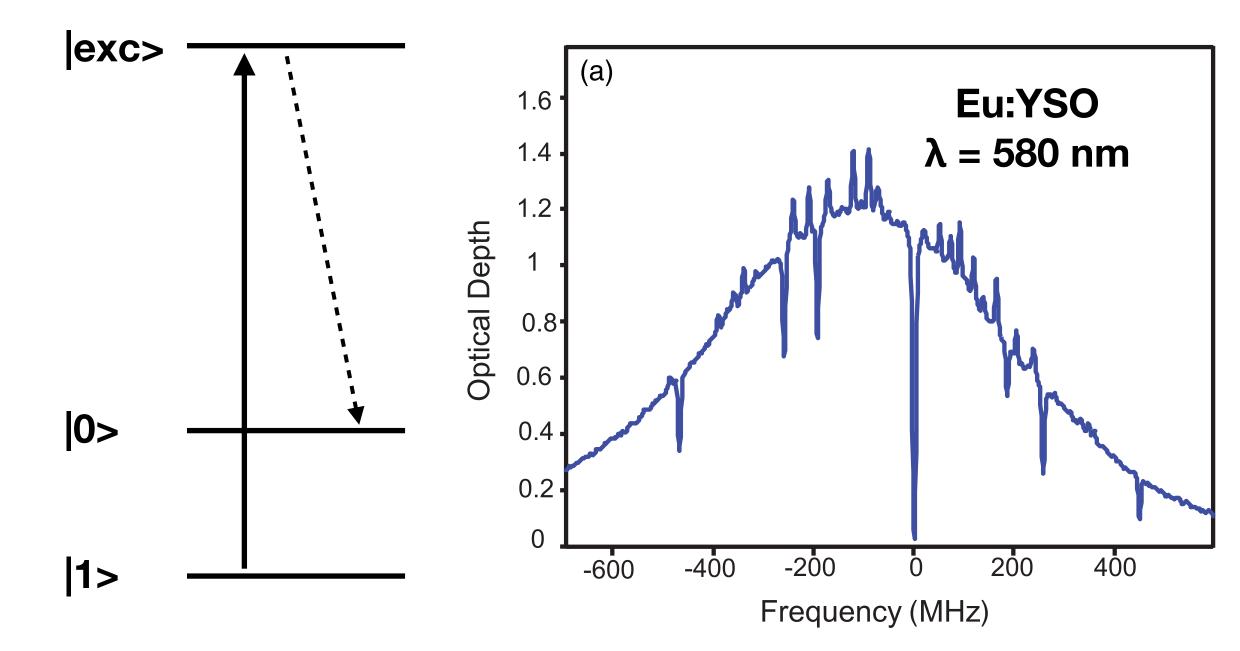
### **High fidelity**

A. Walther et al., PRA, 2015; I. Roos et al., PRA, 2004. J. J. Longdell et al., PRL, 2004.

### Rare Earth Ions as Qubits

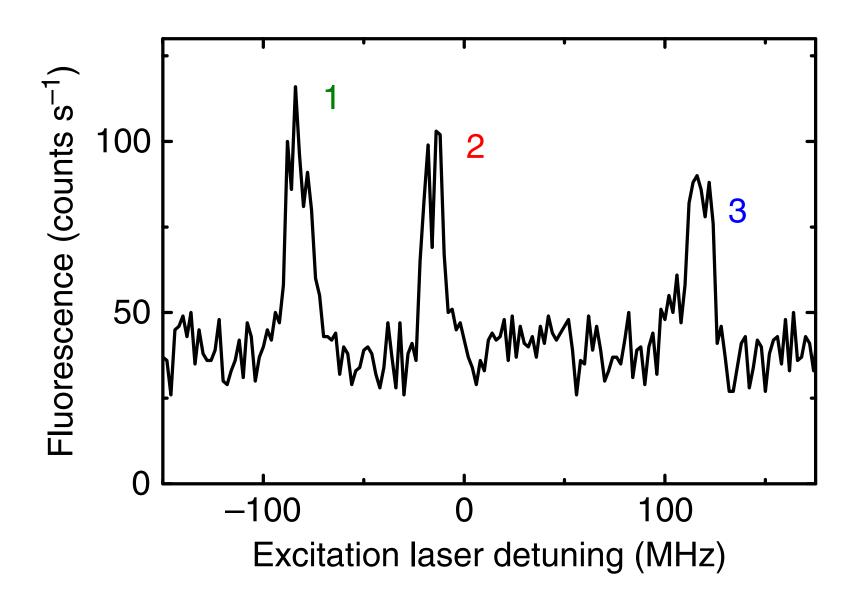
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### Initialization: optical pumping



B. Lauritzen et al., Phys. Rev. B, 2012.

### **Readout:** single ion detection



#### Long T<sub>1</sub>: low count rate

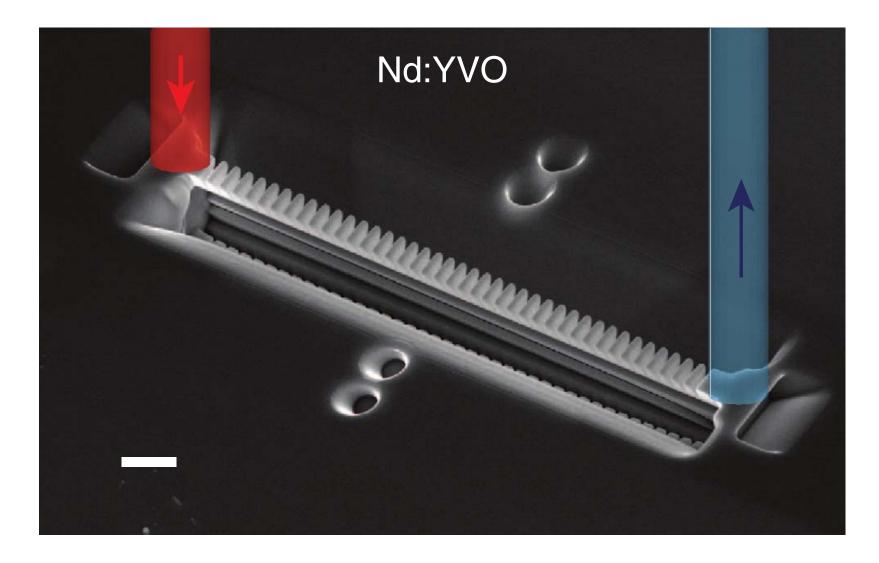
T. Utikal et al. Nat. Commun., 2014; R. Kolesov et al., Nat. Commun. 2012; C. Yin et al., Nature, 2013.

## Improving Detection: Cavities

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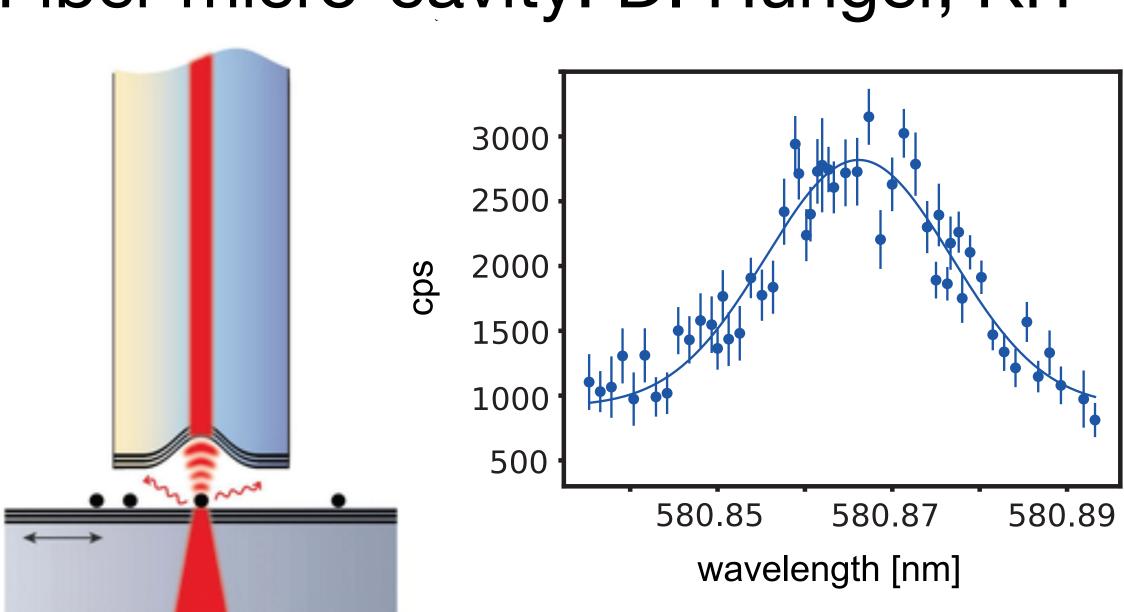
### High Q nano-cavity



### Purcell enhancement High collection efficiency

T. Zhong et al., Nat. Commun., 2015., T. Zhong et al., arXiv, 2018, A. Dibos et al., arXiv, 2017.

### Fiber micro-cavity: D. Hunger, KIT



### High-Q tunable cavity Rare earth nanoparticles

B. Casabone,...,PG, H. de Riedmatten, and D. Hunger, arXiv, 2018.

# Rare earth spins at the nanoscale

## **Optical Spin Control**

exc>

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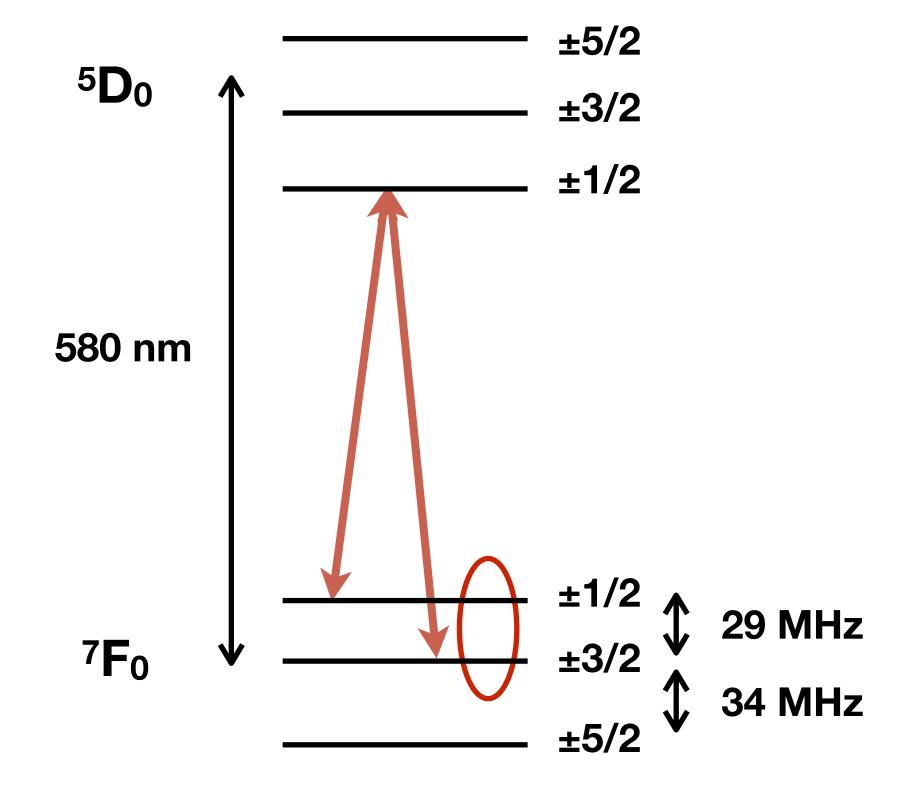
### Extra degree of freedom

Interface w/ photonic qubits Faster operations Easier implementation



G. Waldherr et al., Nature, 2014. D. Press, Nat. Photonics 2010.





J. Karlsson et al. J. Phys.: Condens. Matter 2017.

## Nanoparticles

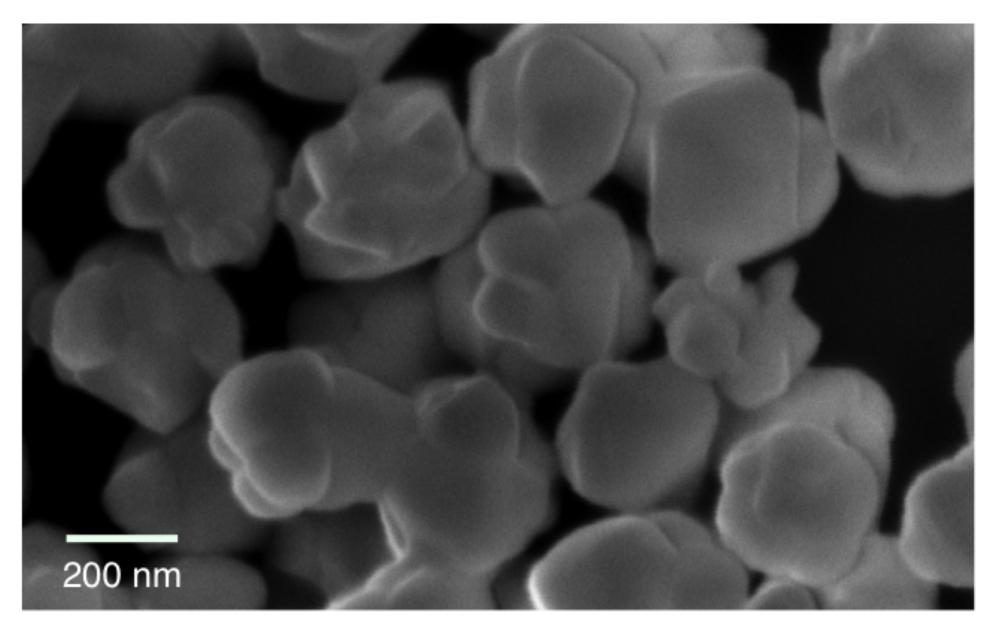
### 0.5% Eu<sup>3+</sup>:Y<sub>2</sub>O<sub>3</sub>

Homogeneous precipitation Monodispersed, spherical

High temperature annealing Cubic phase Defects reduced at 1200 °C

Long optical T<sub>2</sub> in bulk crystal and transparent ceramics

Particles: K. de Oliveira Lima, ..., PG, J. Lumin. 2015.

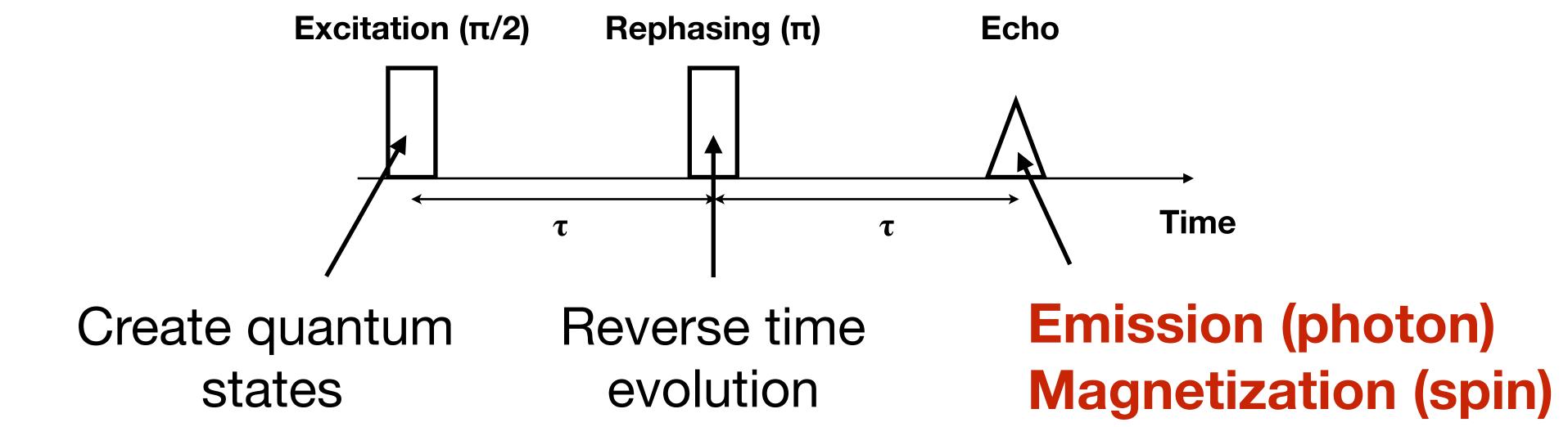


#### Particle size: 400 nm Crystallite size: 130 nm

Ceramics: A. Ferrier, ..., PG, Phys Rev B 2013 - N. Kunkel, ..., PG, APL Mat. 2015, J. Phys. Chem. C 2016, PRB 2017.

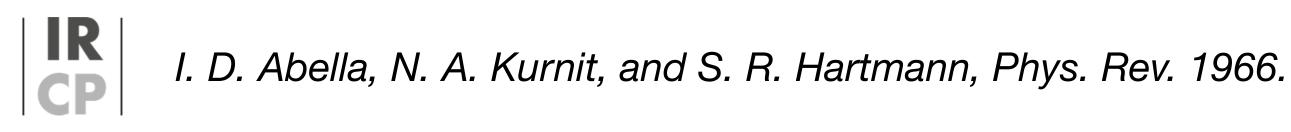


### The Echo Technique

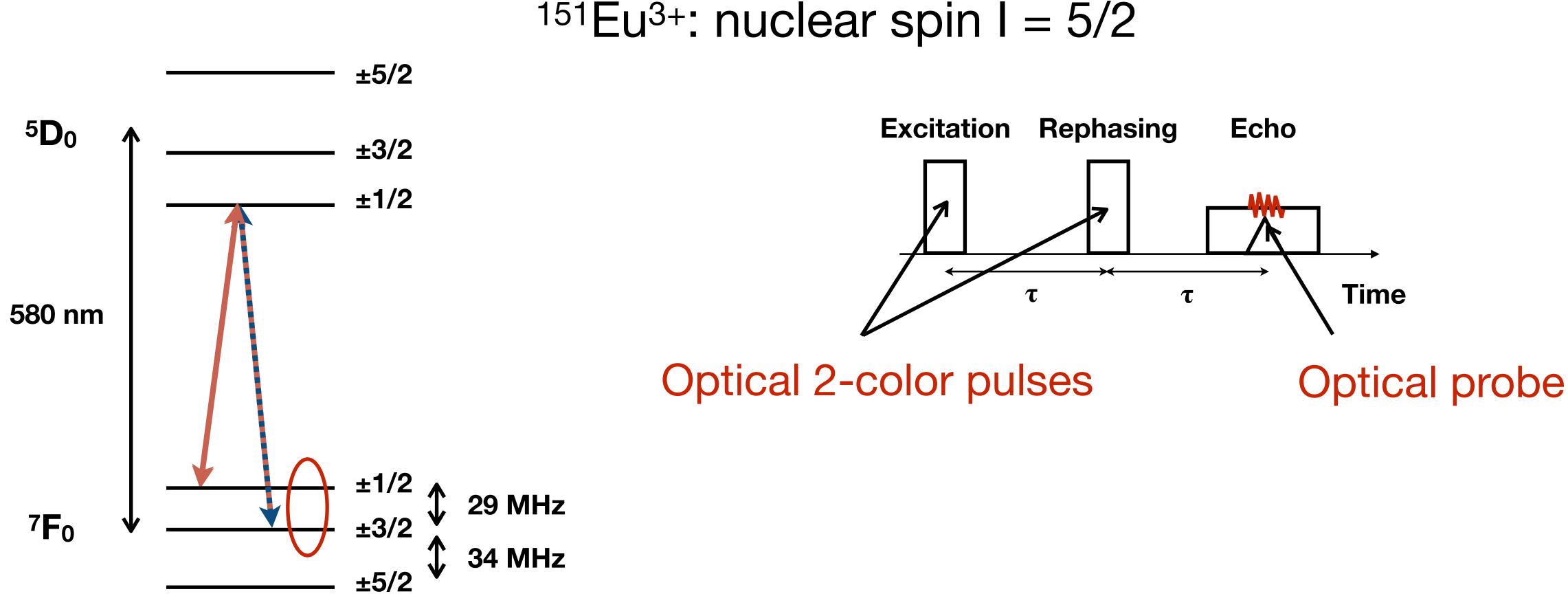


Echo: only ions with unperturbed quantum states Coherence lifetime:  $I_{echo} = exp(-4\tau/T_2)$ Homogeneous linewith:  $\Gamma_h = (\pi T_2)^{-1}$ 

E. L. Hahn, Phys. Rev. 1950.







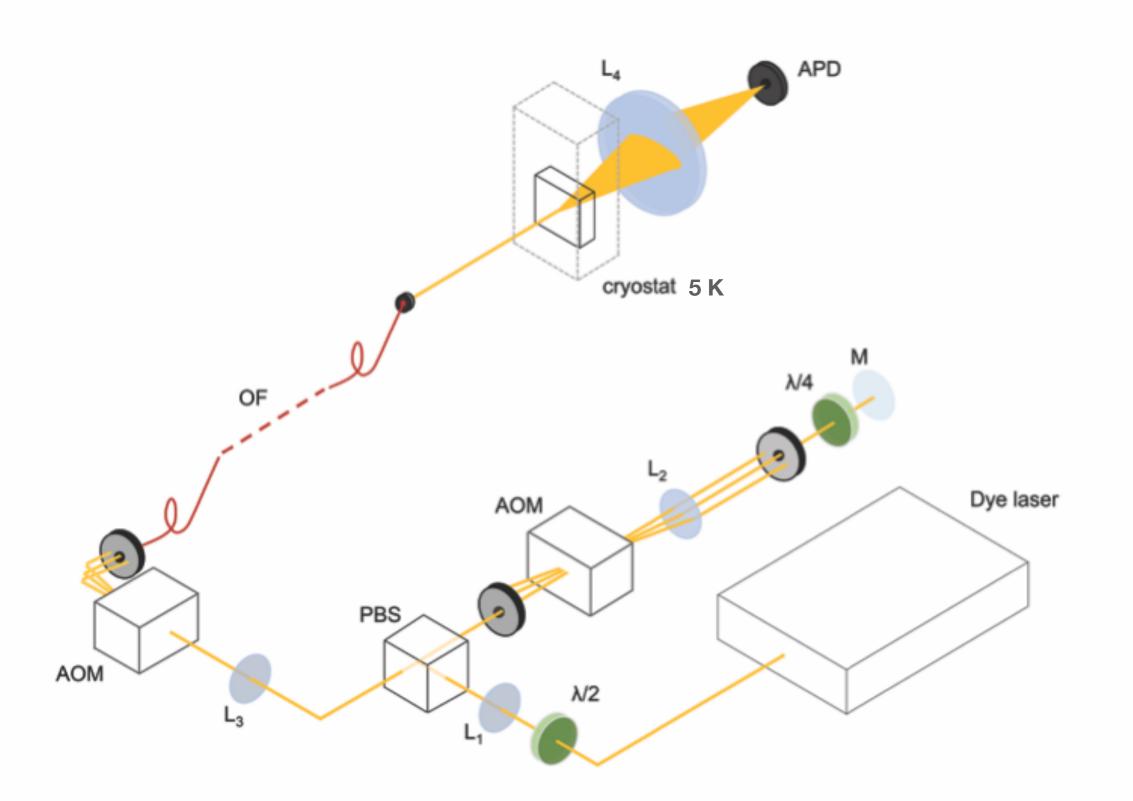
J. Mlynek et al., Phys. Rev. Lett., 1983.

### **All-optical Spin Echoes**

D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and IR PG, arXiv, 2017.

### **Experimental Setup**

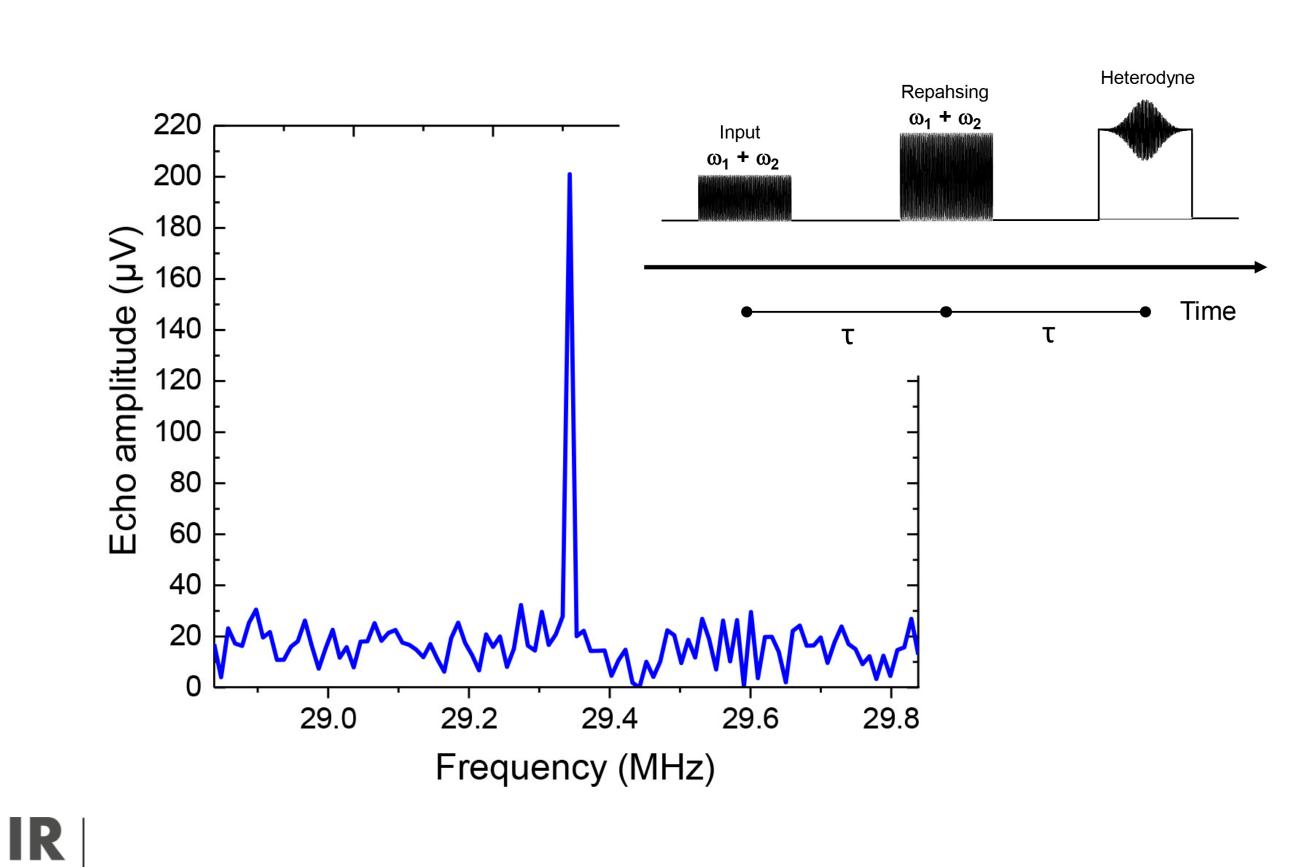
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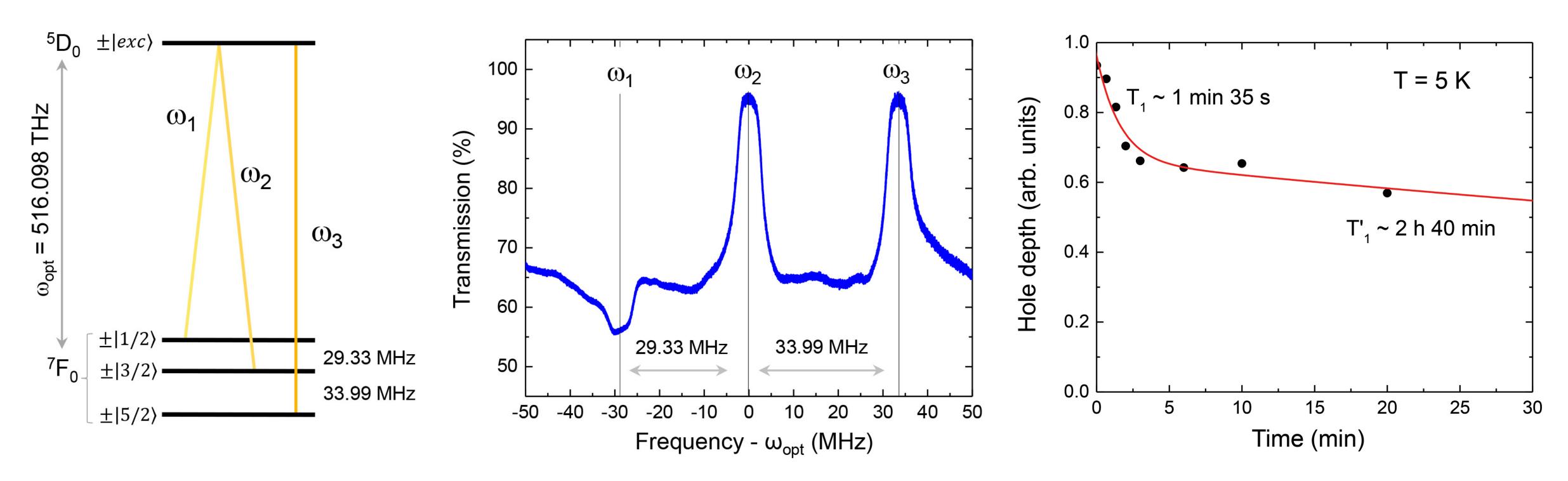
#### Sample: powder!

R. Pierrat et al., arXiv, 2018.

### Echo: FFT of heterodyne signal



### Spin polarization: increased signal



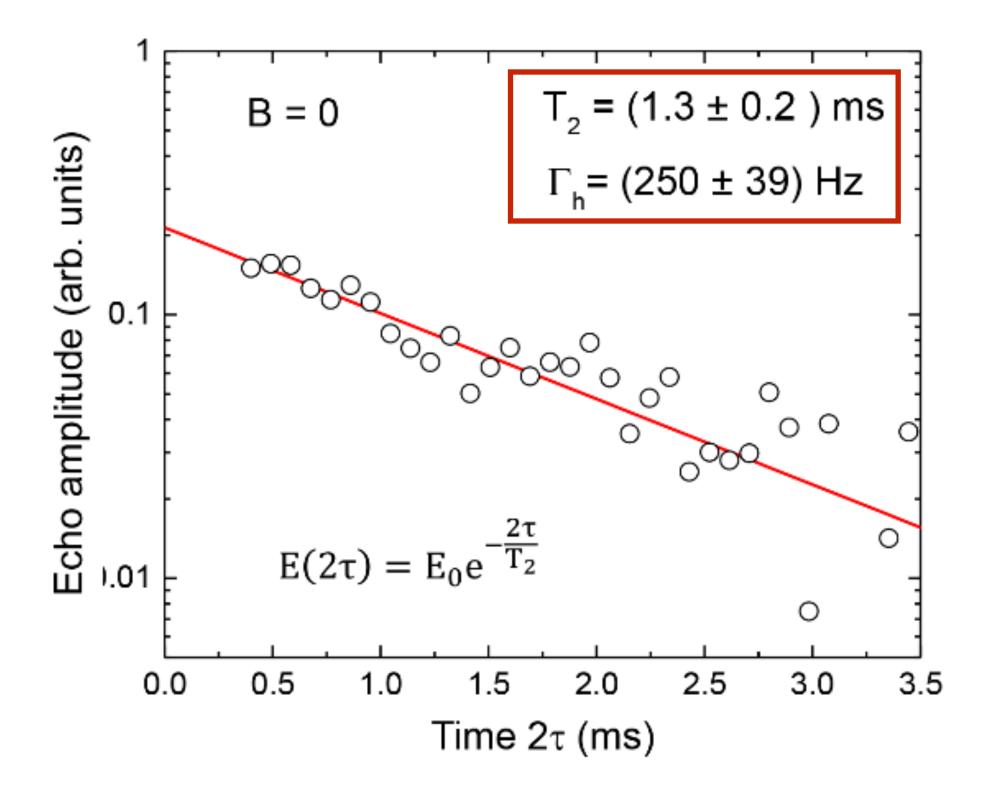
#### D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and PG, arXiv, 2017.

## **Optical Pumping**

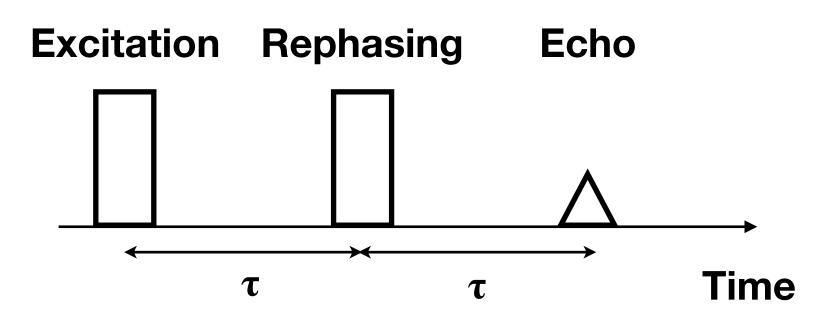
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### Spin Coherence Lifetimes

IR



D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and PG, arXiv, 2017.

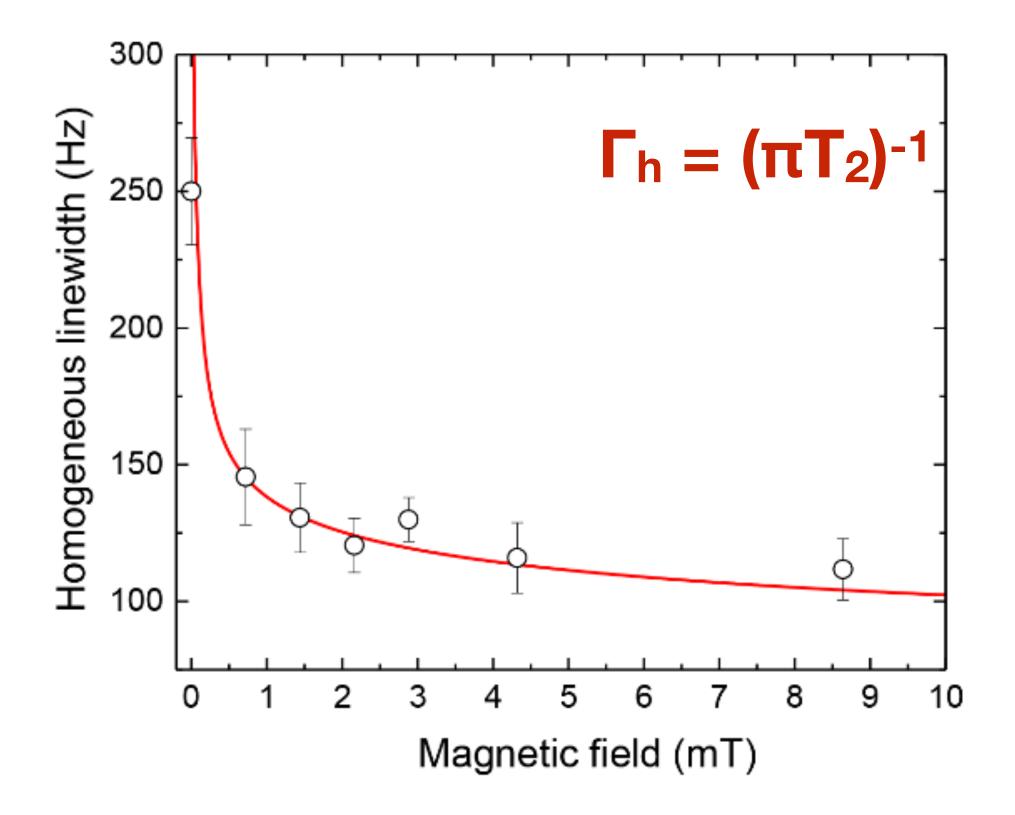


### Spin $T_2$ in the ms range

#### $Eu^{3+}:Y_2O_3$ transparent ceramics: T<sub>2</sub> = 12 ms

#### $Eu^{3+}:Y_2SiO_5$ single crystal: $T_2 = 19$ ms

J. Karlsson, N. Kunkel, A. Ikesue, A. Ferrier, and PG, J. Phys.: Condens. Matter 2017.



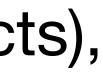
### **Magnetic Field Effects**

### **T**<sub>2</sub> **up to 2.9 ms Strong homogeneous linewidth** decrease for field of 1-2 mT

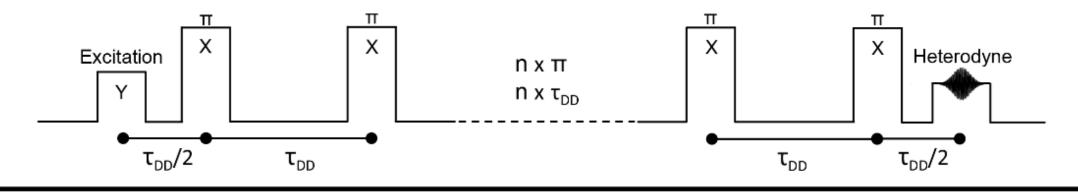
Interactions with electron spins (defects), reduced when H<sub>dd</sub><<H<sub>Zeeman</sub>

### **Longer lifetimes possible** in higher quality samples

IR D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and PG, arXiv, 2017.



#### CPMG sequence



Time

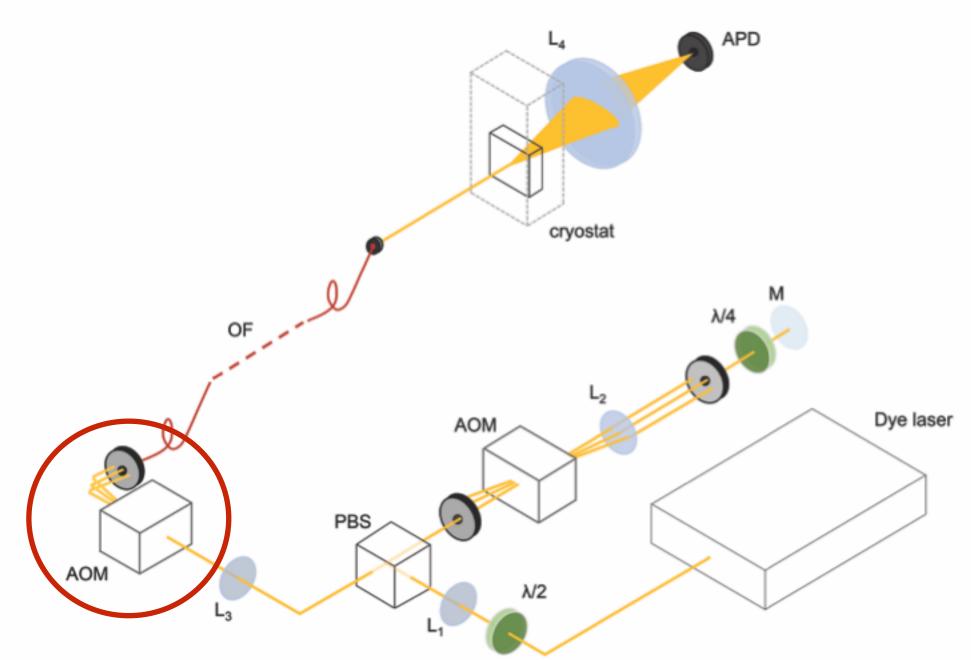
Compensate perturbations τ<sub>c</sub>>>τ<sub>DD</sub>

Efficient for specific initial state

#### **Relative phases are important**

## Dynamical Decoupling

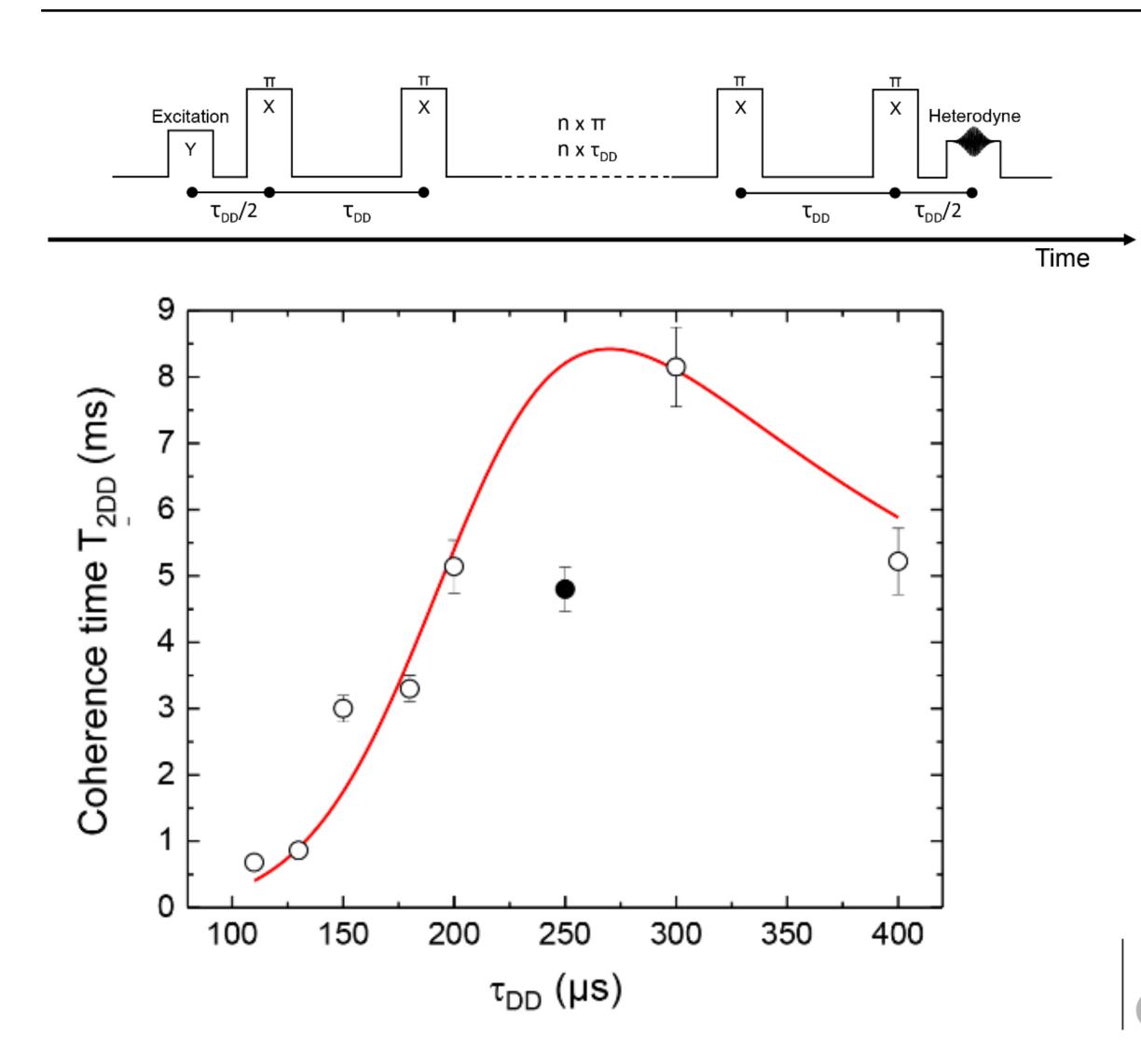
### 2-color pulses: single AOM



#### **High phase stability over minutes**

IR D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and PG, arXiv, 2017.

### T<sub>2</sub> Extension



#### T<sub>2</sub> up to 8 ms

Balance between decoupling and pulse error accumulation

Pulse fidelity: limited by the strong scattering in the powder

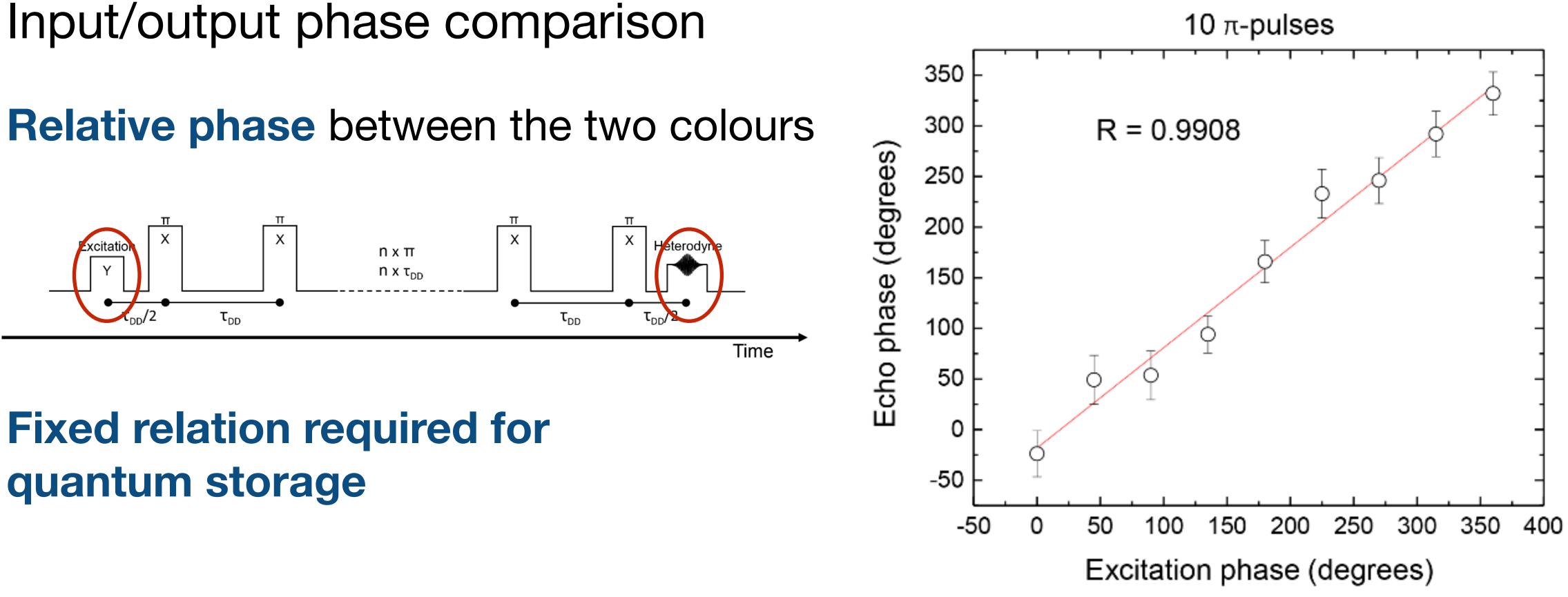
Larger decoupling in a single particle

IR D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and *PG, arXiv, 2017.* 



### Phase Coherence

### Input/output phase comparison



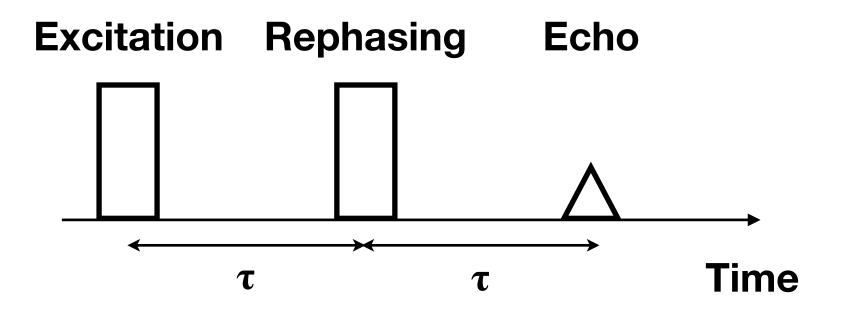
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**Fixed relation required for** quantum storage

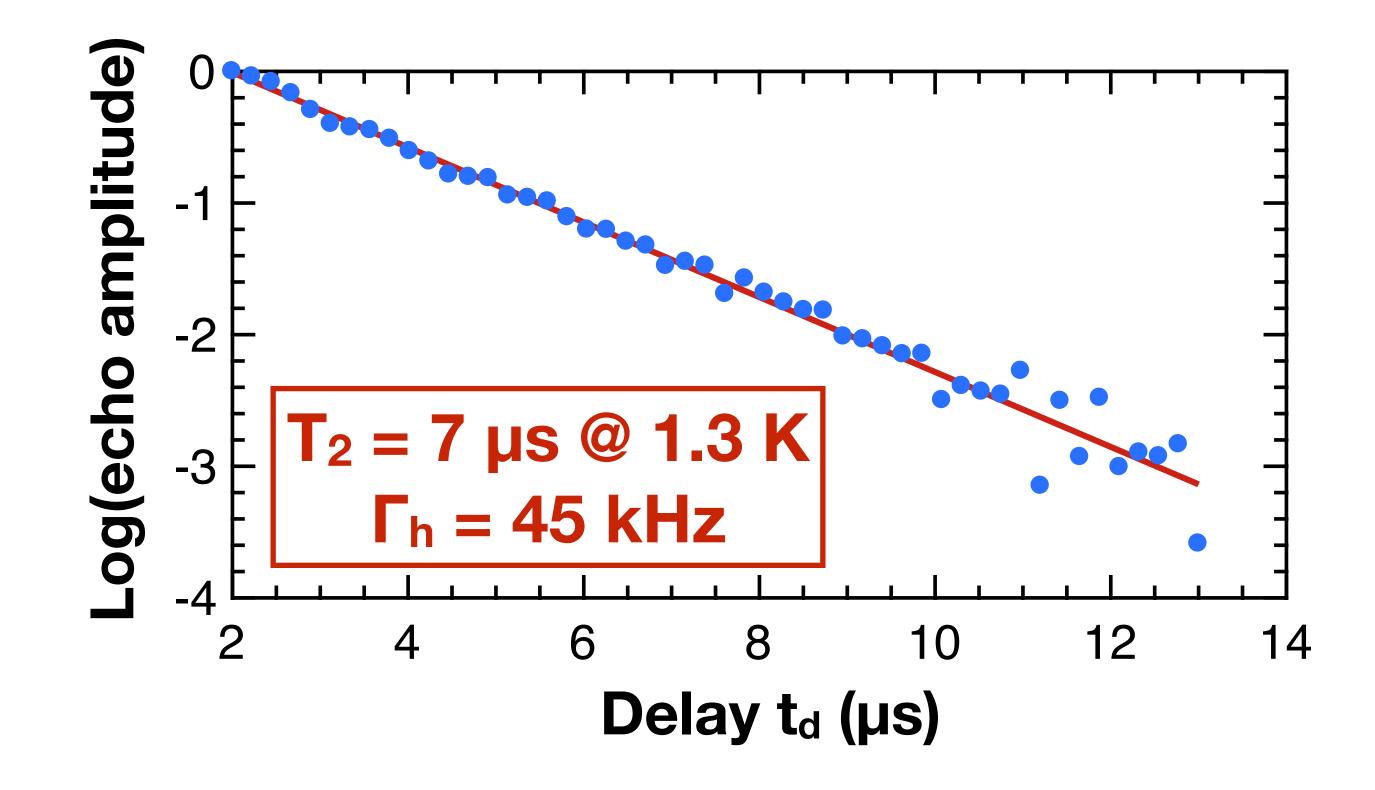
> D. Serrano, J. Karlsson, A. Fossati, A. Ferrier, and *PG, arXiv, 2017.*

## Optical T<sub>2</sub> in nanocrystals

### Echo Decay in Nanocrystals



### Suitable for high fidelity spin gates



IR J. G. Bartholomew, K. de Oliveira Lima, A. Ferrier, and PG, Nano. Lett. 2017.

## Acknowledgment

Paris team:

A. Ferrier, D. Serrano, A. Tallaire, M. Mortier, Shuping Liu, Zhonghan Zhang, Sacha Welinski, Alexandre Fossati, Marion Scarafagio. Former members: Marko Lovrić, Karmel de Oliveira Lima, John Bartholomew, **Jenny Karlsson** 

Collaborators: R. Gonçalves, USP, Brazil - D. Hunger, KIT, Germany - S. Kröll, Lund University, Sweden Y. Le Coq, SYRTE, France - S. Seidelin, Grenoble University, France - H. de Riedmatten, ICFO, Spain - F. Koppens, ICFO, Spain - K. Mølmer, Aarhus University, Denmark N. Oliverio, Keysight Inc., USA



Funding:

### JanOQTech

Nanoscale Systems for Optical Quantum Technologies http://www.nanogtech.eu

European Union's Horizon 2020 programme





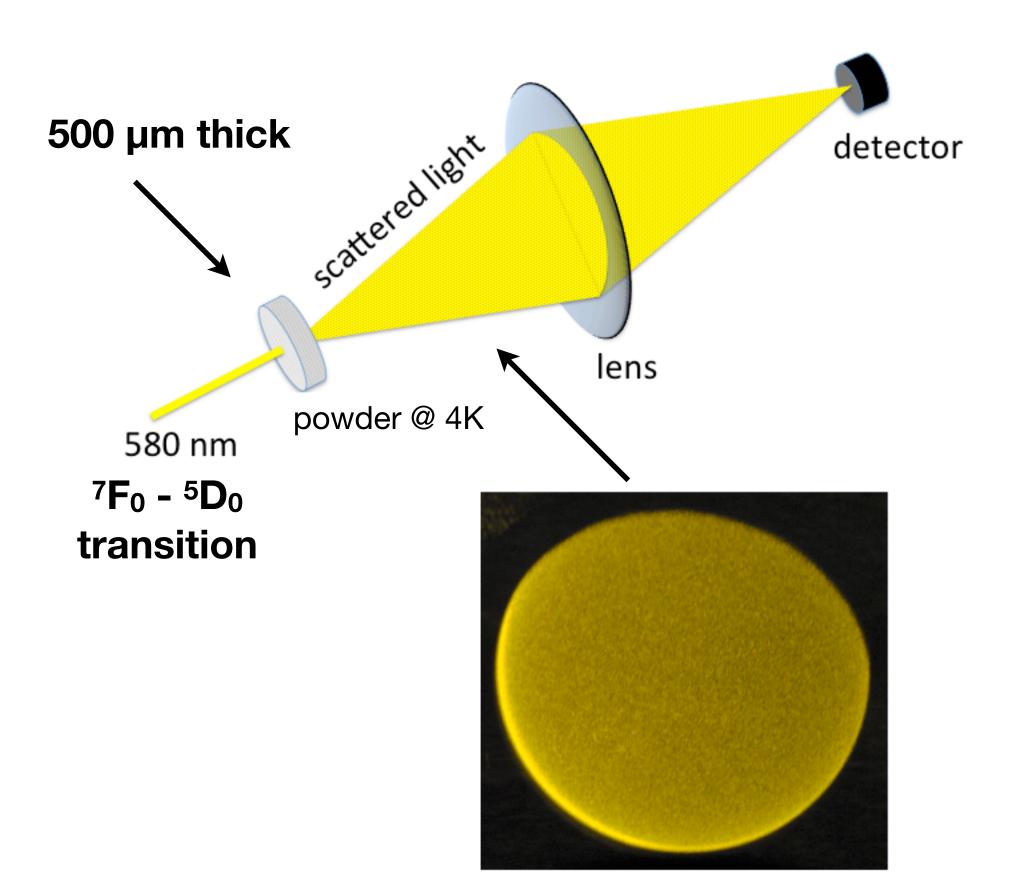


## Summary

- Coherence lifetime extension by dynamical decoupling using trains of 2-color optical pulses
- T<sub>2</sub> improvement by lowering magnetic defects concentration and in single particles
- Rare earth doped nanoparticles: optically controlled qubits

Nuclear spins with ms long coherence lifetimes measured by all-optical techniques

### Photon Echo in Powders



#### Light scattered by the powder

A. Perrot, PG, et al. Phys. Rev. Lett. 2013. F. Beaudoux, ..., PG, Opt. Express 2011.

### Interferometric detection

