



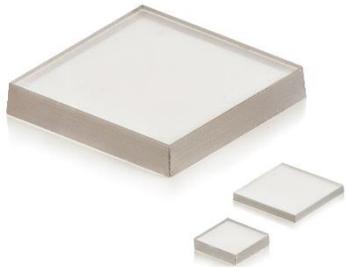
# Group-IV defects in diamond for quantum networks

Petr Siyushev

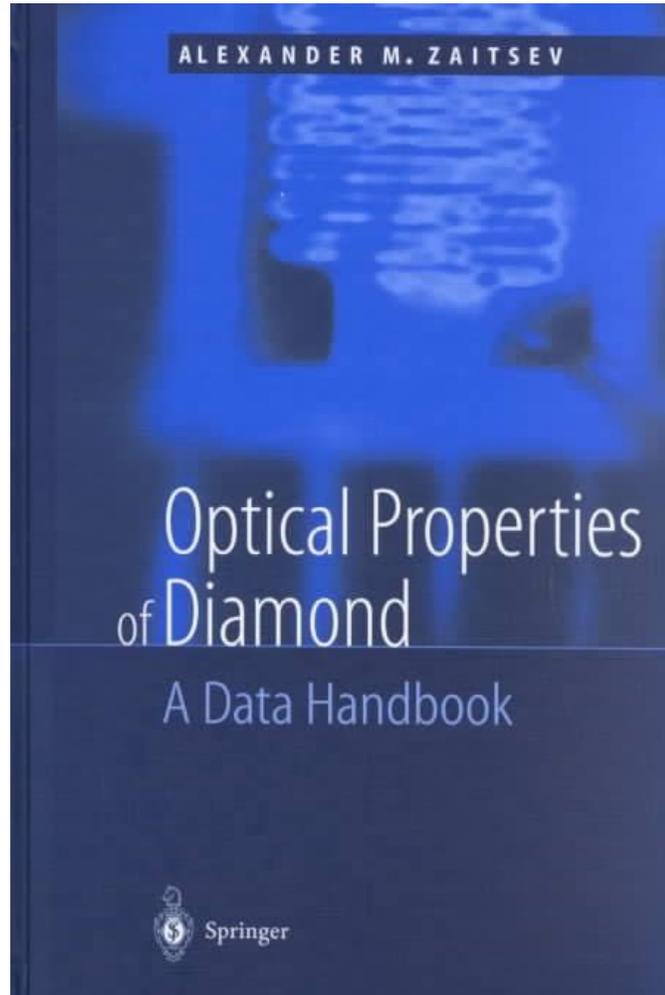
*Institute for Quantum Optics, University of Ulm*



# Color centers in diamond



Boring electronic grade diamonds

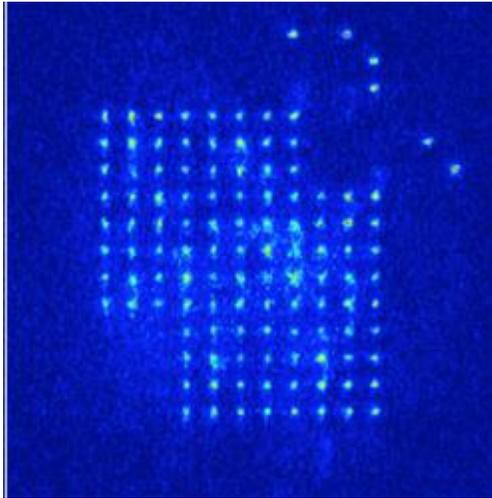


Besides **Nitrogen** and **Boron**, more than 500 defects are known!

but structurally only about 20 are explored

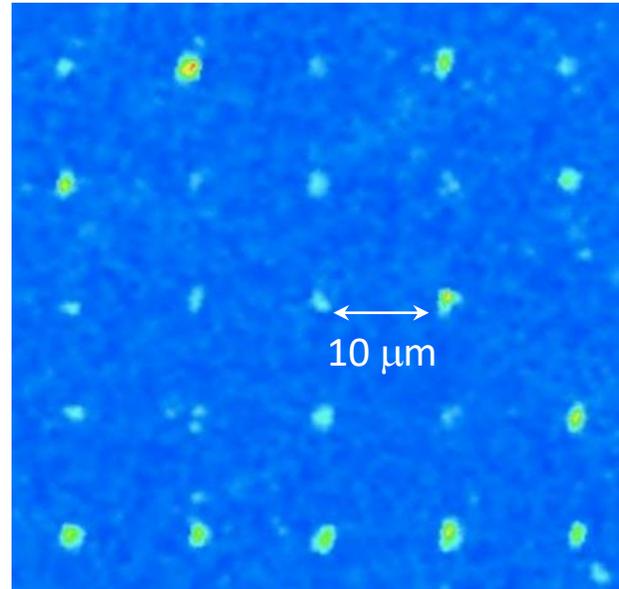
# Color centers in diamond

Trapped atoms



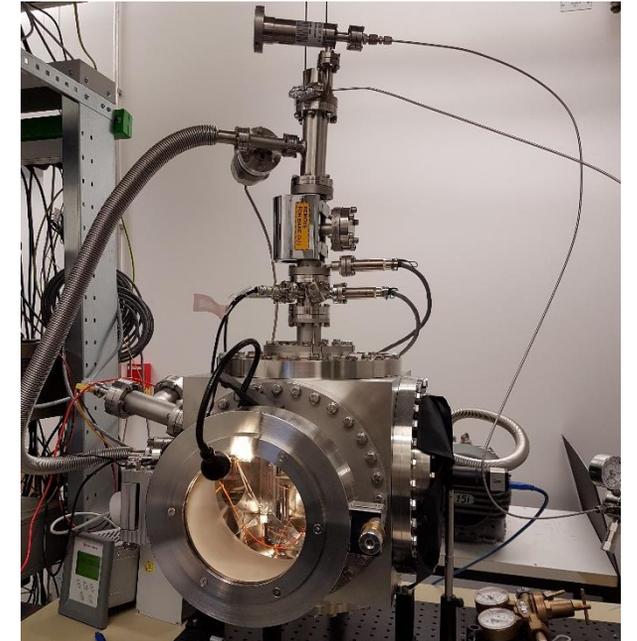
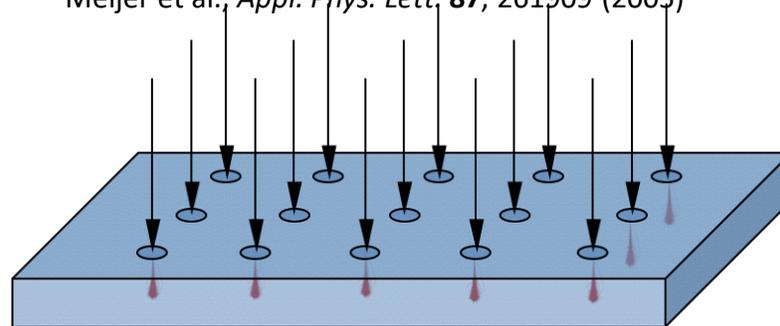
D. Ohi de Mello et al., *Phys. Rev. Lett.* **122**, 203601 (2019)

“Trapped” color centers in crystal



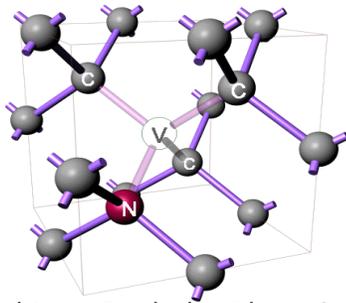
Meijer et al., *Appl. Phys. Lett.* **87**, 261909 (2005)

- ✓ Well isolated
- ✓ Well controllable
- × Difficult to trap and store



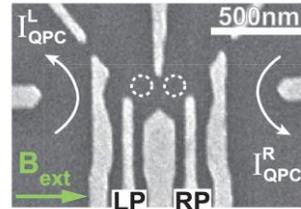
- ✓ Defects are atomic like systems
- ✓ Intrinsically trapped in solids
- ✓ Full quantum control
- ✓ Potentially scalable

# Solid-state spin qubits



*J. Wrachtrup, F. Jelezko, Phys.: Condens. Matter* **16**, R1089–R1104 (2004)

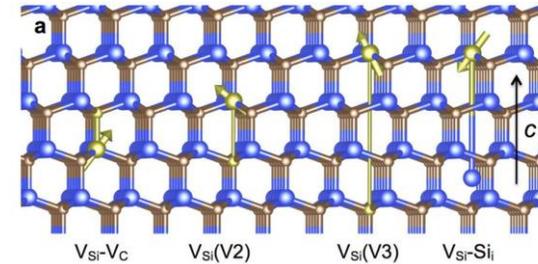
NV centers in diamond



*K. C. Nowack et al. Science*

**333**, 1269–1272 (2011)

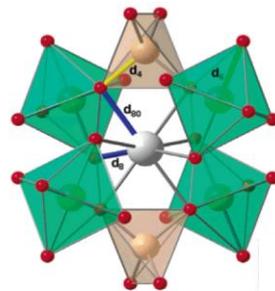
Quantum dots



*H. Kraus et al. Sci. Rep.* **4**, 5303 (2014)

$V_{Si}$ ,  $V_{Si-V_C}$  in SiC

Solid-state spin qubits



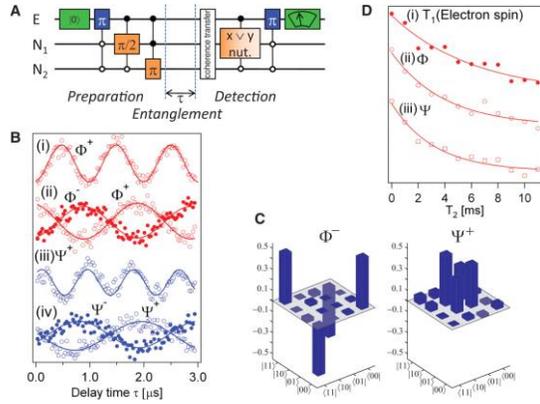
*P. Siyushev et al. Nat. Commun.* **4**, 3895 (2014)

Rare-earth ions

Others...

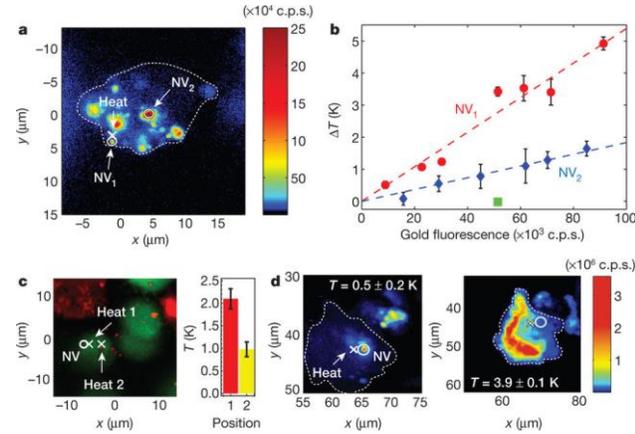
# can be used as

## Qubit



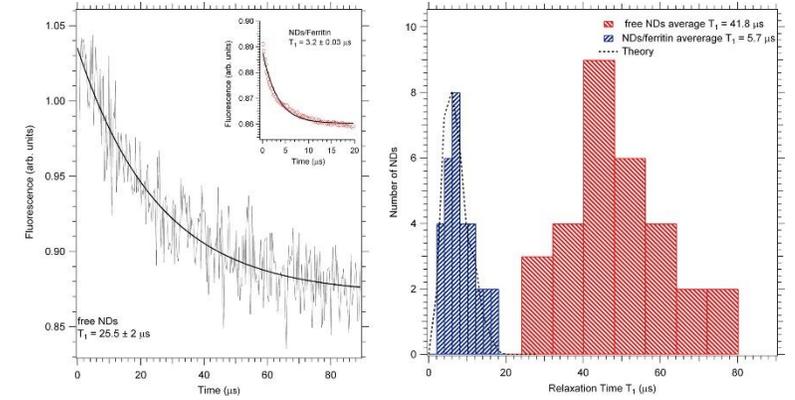
*P. Neumann et al., Science 320, 1326-1329 (2008)*

## Temperature sensor



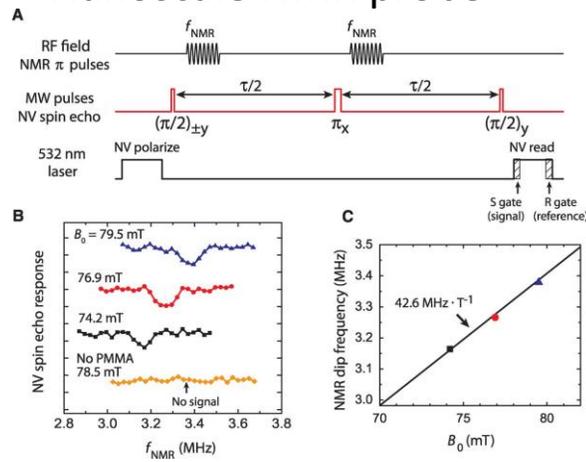
*G. Kucsko et al., Nature 500, 54-58 (2013)*

## Magnetic sensor



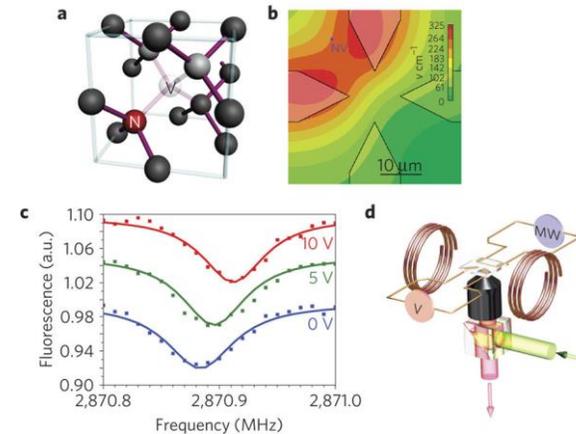
*A. Ermakova et al., Nano Lett. 13, 3305-3309 (2013)*

## Nanoscale NMR probe



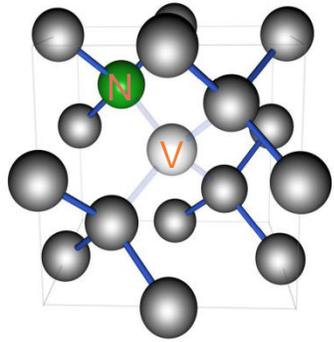
*H. J. Mamin et al., Science 339, 557-560 (2013)*

## Electric field sensor



*F. Dolde et al., Nat. Phys. 7, 459-463 (2011)*

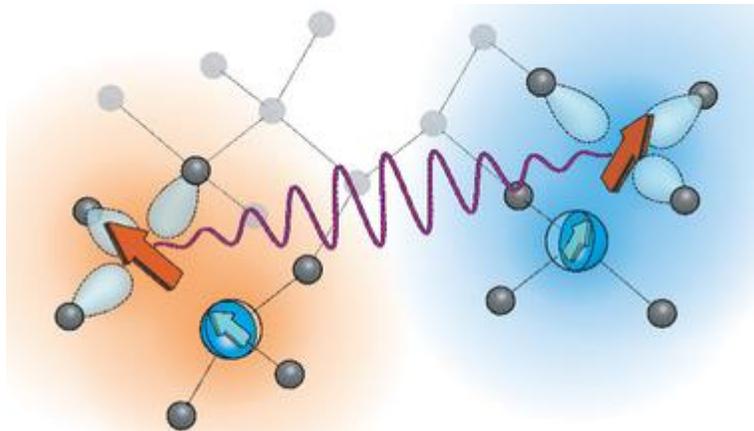
# Why searching for new defects?



Optical initialization and readout

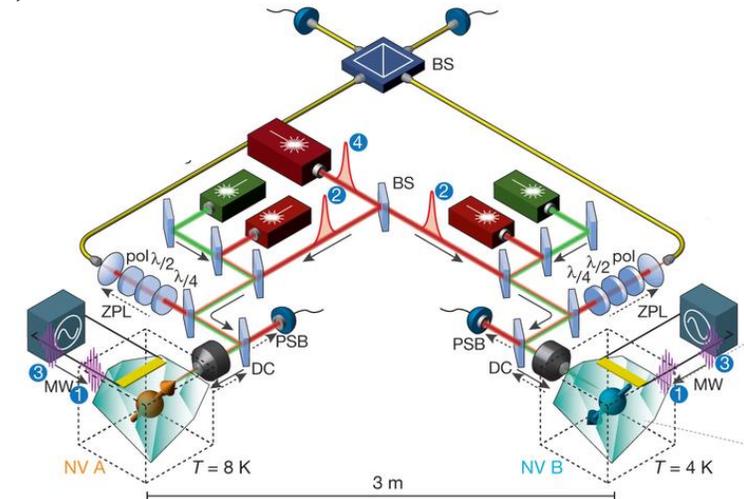
Long spin coherence time

Spin manipulation by MW field



Entanglement between two NV centers

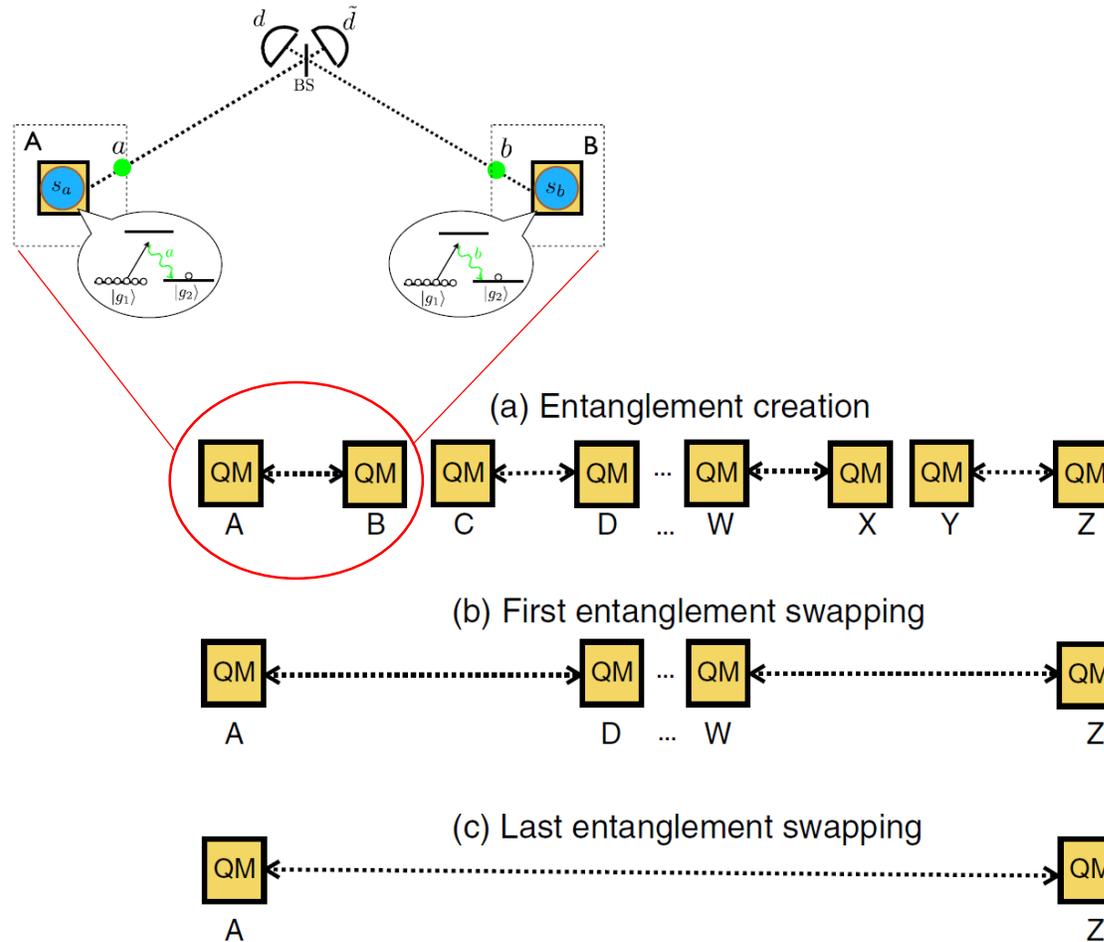
*F. Dolde et al., Nat. Phys. 9, 139–143 (2013)*



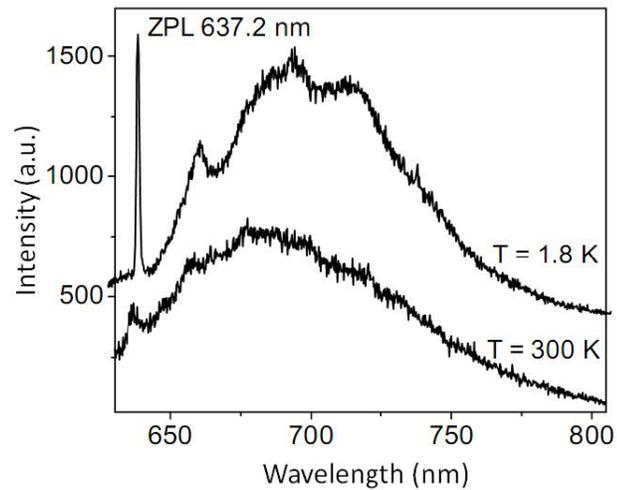
*H. Bernien et al., Nature 497, 86–90 (2013)*

One entanglement event per 10 minutes!

# Spin-photon interface is essential...

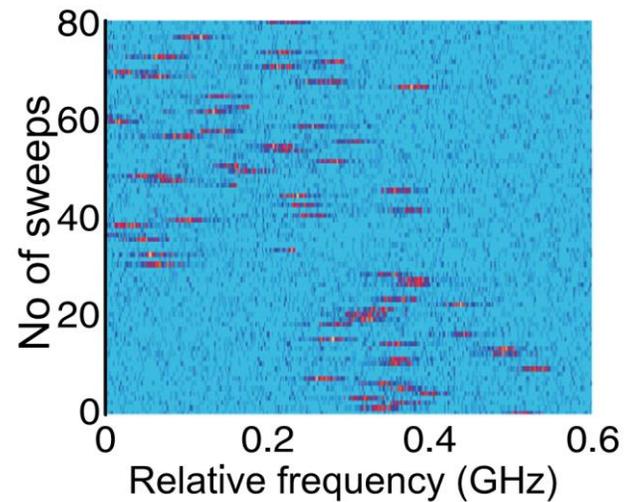


# Entanglement rate is poor?



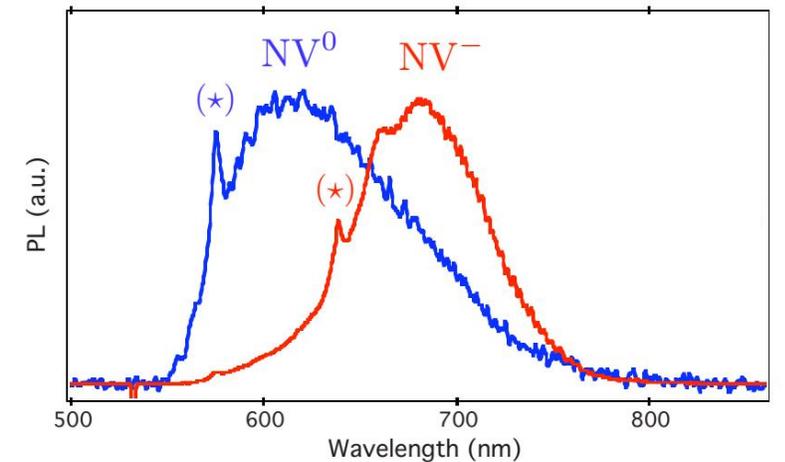
*F. Jelezko and J. Wrachtrup, Phys. Stat. Solidi* **204** 3207 (2006)

Low Debye-Waller factor  $\sim 0.4$



*P. Siyushev et al., Phys. Rev. Lett.* **110**, 167402 (2013)

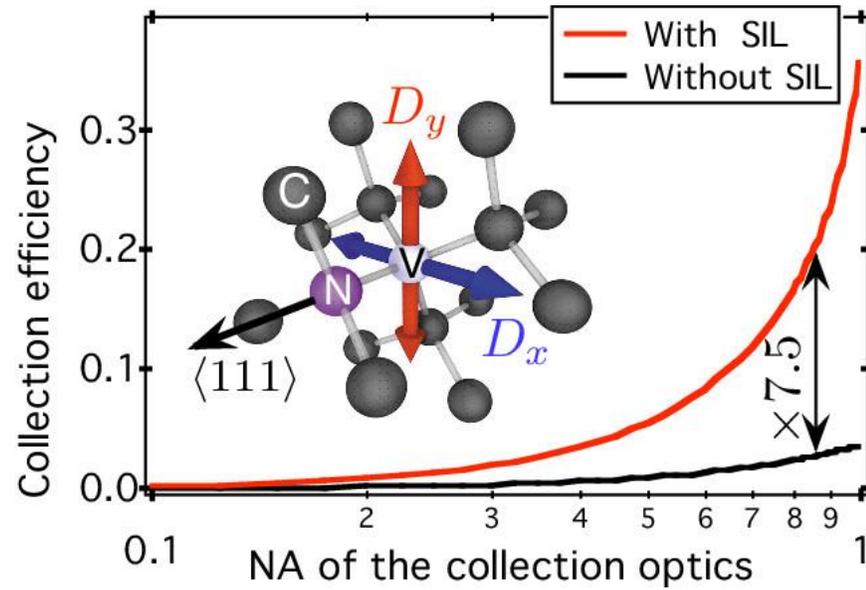
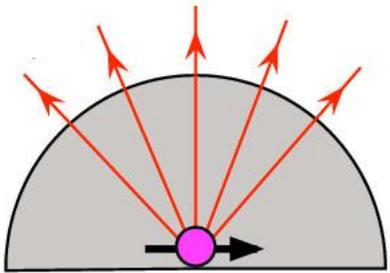
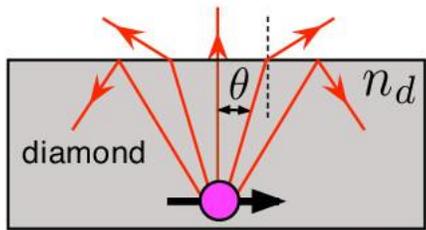
Spectral jumps



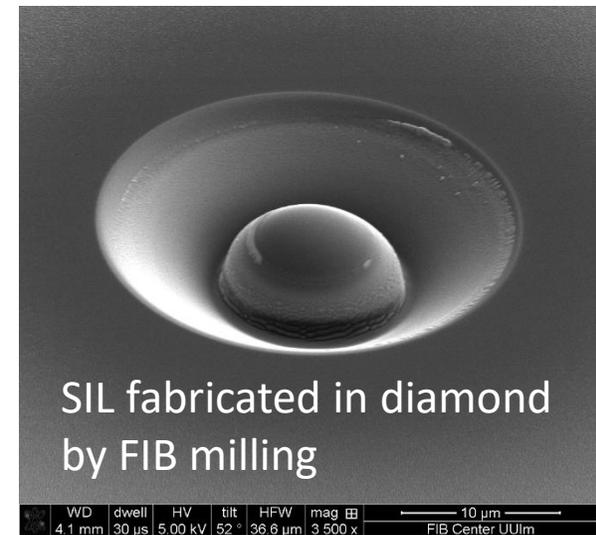
*L. Rondin et al., Phys Rev. B* **82**, 115449 (2010)

Emission from NV<sup>0</sup>

# Collection efficiency enhancement



P. Siyushev et al., *Appl. Phys. Lett.* **97**, 241902 (2010)



# Other candidates...

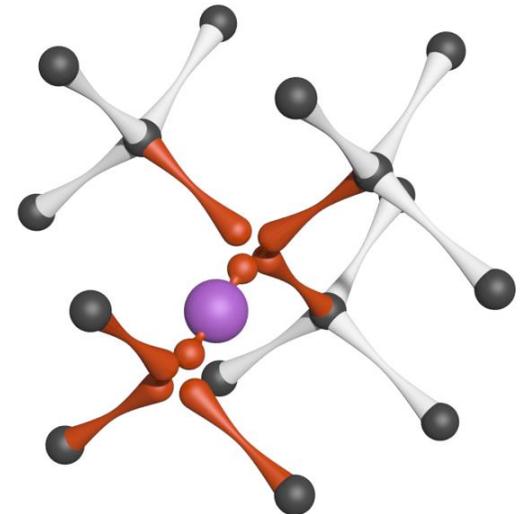
		Group																							
		a	I	b	a	II	b	a	III	b	a	IV	b	a	V	b	a	VI	b	a	VII	b	a	VIII	b
1	I	1 H																						2 He	
2	II	3 Li		4 Be		5 B		6 C		7 N		8 O		9 F		10 Ne									
3	III	11 Na		12 Mg		13 Al		14 Si		15 P		16 S		17 Cl		18 Ar									
4	IV	19 K		20 Ca		21 Sc		22 Ti		23 V		24 Cr		25 Mn		26 Fe	27 Co	28 Ni							
	V	29 Cu		30 Zn		31 Ga		32 Ge		33 As		34 Se		35 Br		36 Kr									
5	VI	37 Rb		38 Sr		39 Y		40 Zr		41 Nb		42 Mo		43 Tc		44 Ru	45 Rh	46 Pd							
	VII	47 Ag		48 Cd		49 In		50 Sn		51 Sb		52 Te		53 I		54 Xe									
6	VIII	55 Cs		56 Ba		57-71		72 Hf		73 Ta		74 W		75 Re		76 Os	77 Ir	78 Pt							
	IX	79 Au		80 Hg		81 Tl		82 Pb		83 Bi		84 Po		85 At		86 Rn									
7	X	87 Fr		88 Ra		89-103		104 Rf		105 Db		106 Sg		107 Bh		108 Hs	109 Mt	110 Ds							
	XI	111 Rg		112 Cn		113 Nh		114 Fl		115 Mc		116 Lv		117 Ts		118 Og									
Higher oxides		R <sub>2</sub> O	RO	R <sub>2</sub> O <sub>3</sub>	RO <sub>2</sub>	R <sub>2</sub> O <sub>5</sub>	RO <sub>3</sub>	R <sub>2</sub> O <sub>7</sub>	RO <sub>4</sub>																
Volatile hydrogen compounds				[(RH <sub>3</sub> ) <sub>x</sub> ]	RH <sub>4</sub>	RH <sub>3</sub>	RH <sub>2</sub>	RH																	
57	58	59	60	61	62	63	64	65	66	67	68	69	70	71											
La	Ce	Pr	Nd	Pm	Sm	Eu	Gd	Tb	Dy	Ho	Er	Tm	Yb	Lu											
89	90	91	92	93	94	95	96	97	98	99	100	101	102	103											
Ac	Th	Pa	U	Np	Pu	Am	Cm	Bk	Cf	Es	Fm	Md	No	Lr											

14 **Si** 2  
Silicon 8  
4

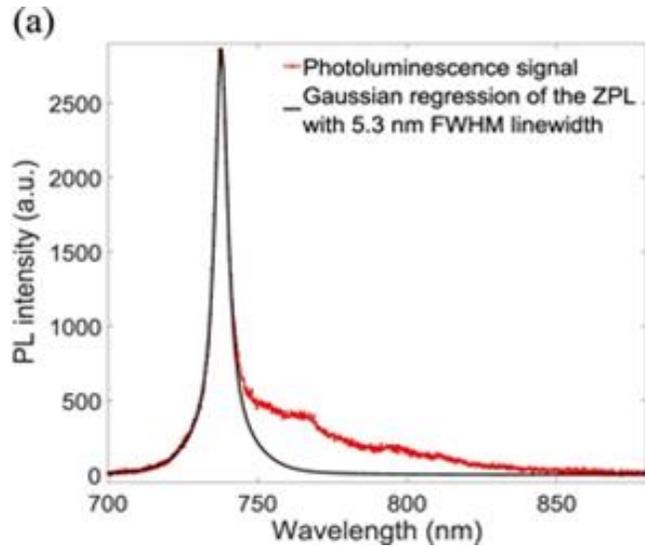
32 **Ge** 2  
Germanium 8  
18  
4

50 **Sn** 2  
Tin 8  
18  
4

82 **Pb** 2  
Lead 8  
18  
32  
4



# Other candidates...



S. Häußler et al. *New J. Phys.* **19** 063036 (2017)

C. Wang et al.

*J. Phys. B: At., Mol. Opt. Phys.* **39**, 37 (2006)

C. Hepp et al. *Phys. Rev. Lett.* **112**, 036405 (2014)

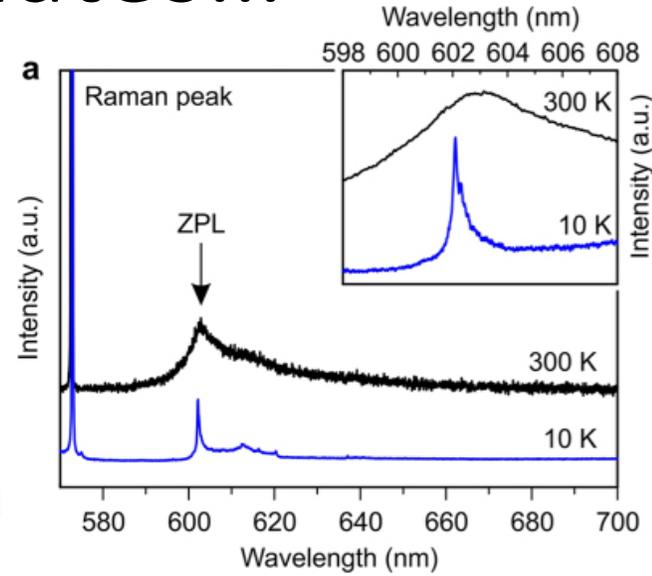
L. Rogers et al. *Phys. Rev. B* **89**, 235101 (2014)

A. Sipahigil et al.

*Phys. Rev. Lett.* **113**, 113602 (2014)

**Many others...**

737 nm



T. Iwasaki et al. *Sci. Rep.* **5**, 12882 (2015)

Y. N. Palyanov et al. *Sci. Rep.* **5**, 14789 (2015)

E. A. Ekimov et al. *JETP Lett.* **102**, 701 (2016)

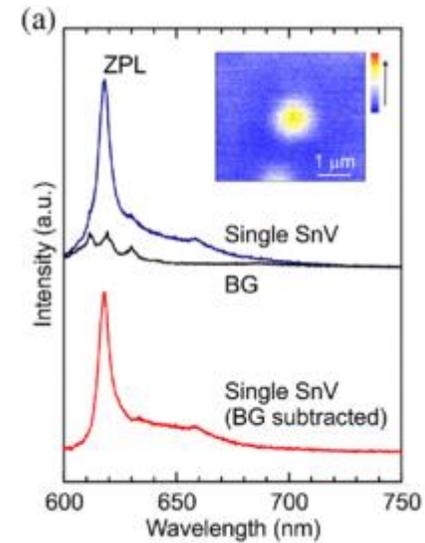
P. Siyushev et al. *Phys. Rev. B* **96**, 081201 (2017)

M. Bhaskar et al.

*Phys. Rev. Lett.* **118**, 223603 (2017)

**Others...**

602 nm



T. Iwasaki et al.

*Phys. Rev. Lett.* **119**, 253601 (2017)

S. Ditalia Tchernij et al.

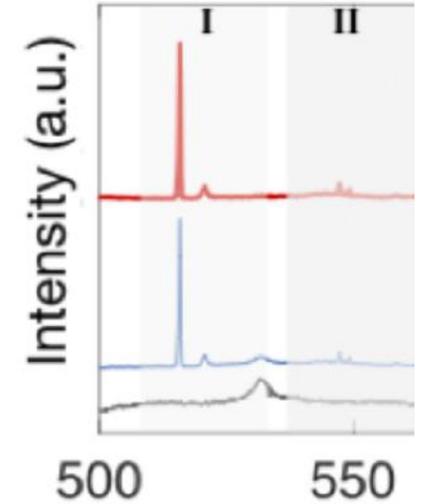
*ACS Photonics* **4**, 2580 (2017)

M. Alkahtani et al.

*Appl. Phys. Lett.* **112**, 241902 (2018)

M. Trusheim et al. arXiv:1811.07777

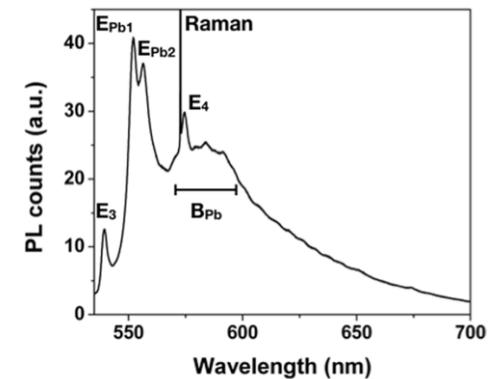
619 nm



M. Trusheim et al.

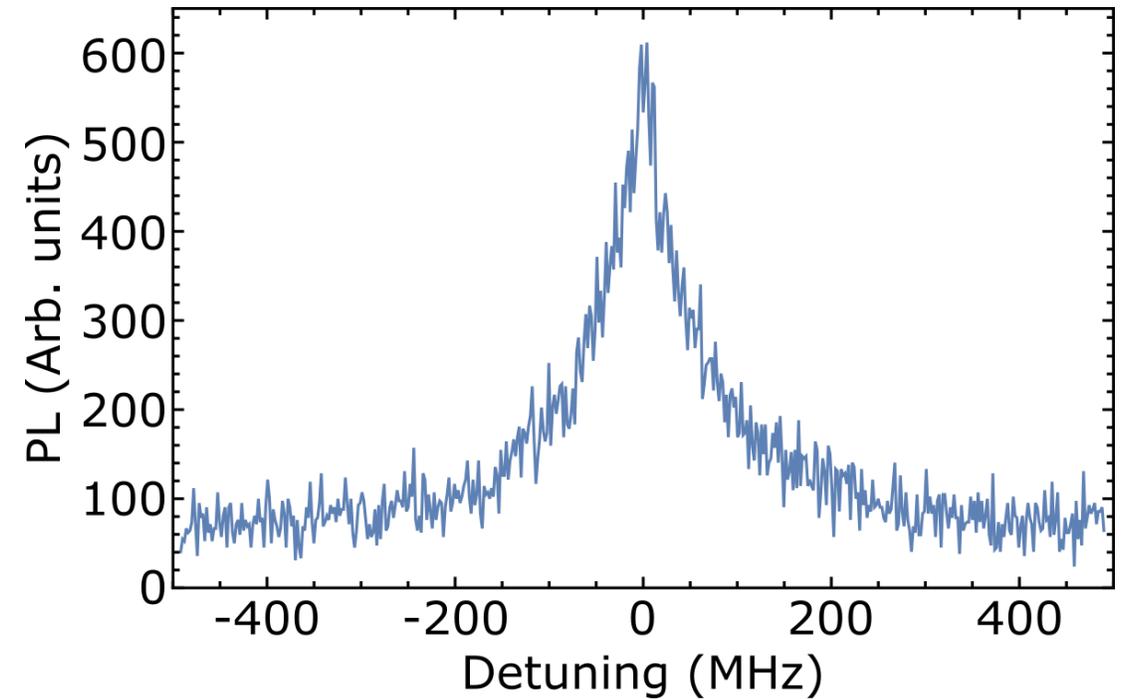
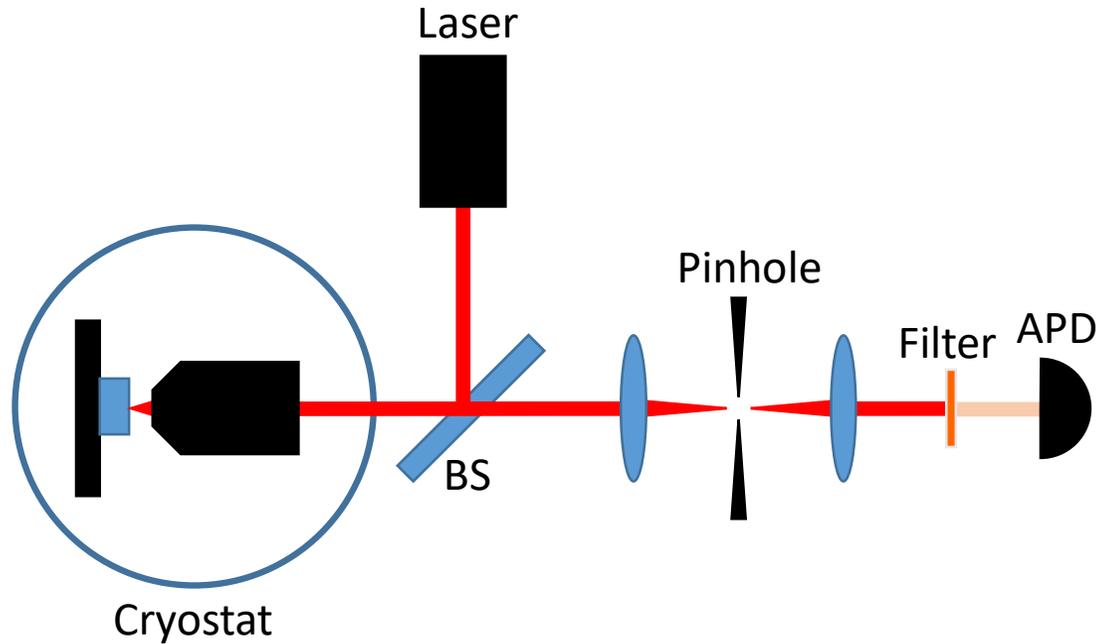
*Phys. Rev. B* **99**, 075430 (2019)

**OR**



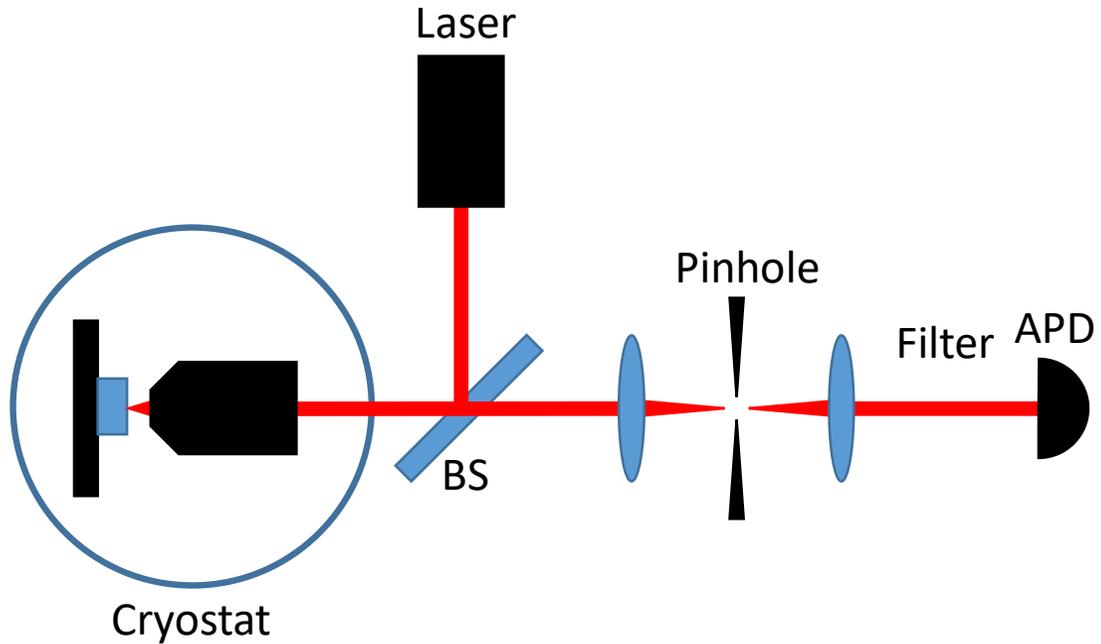
S. Ditalia Tchernij et al, *ACS Photonics* **5**, 4864 (2018)

# Why so special?

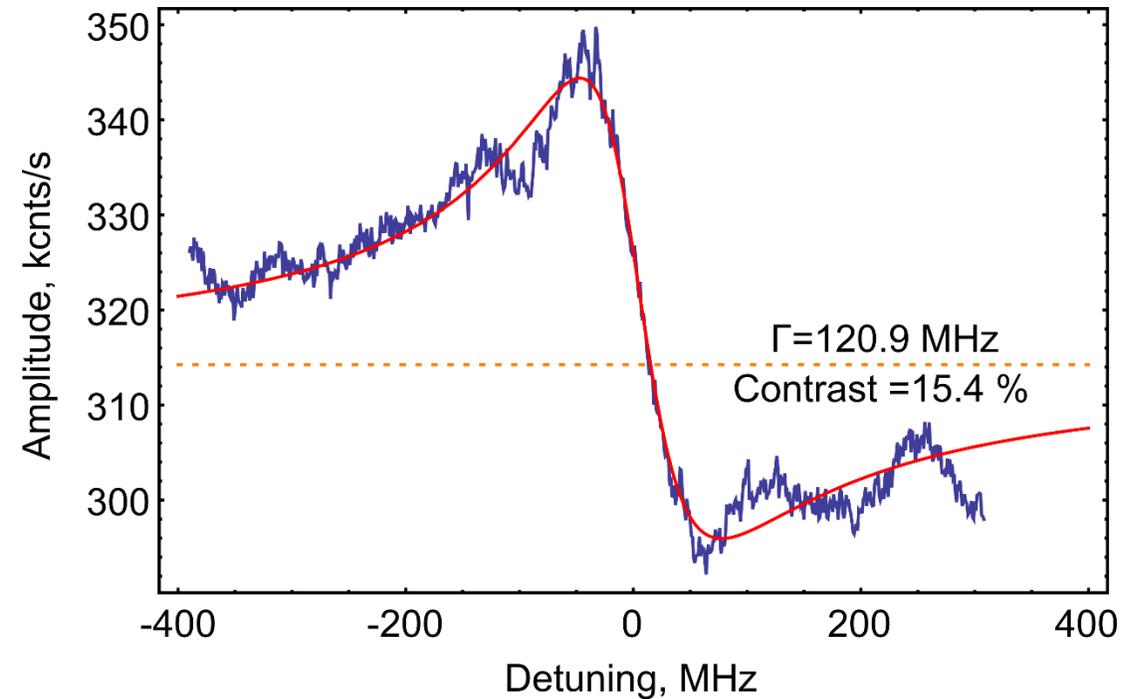


Laser is scanned over ZPL, photons are detected from the PSB  
- photoluminescence excitation spectroscopy (PLE)

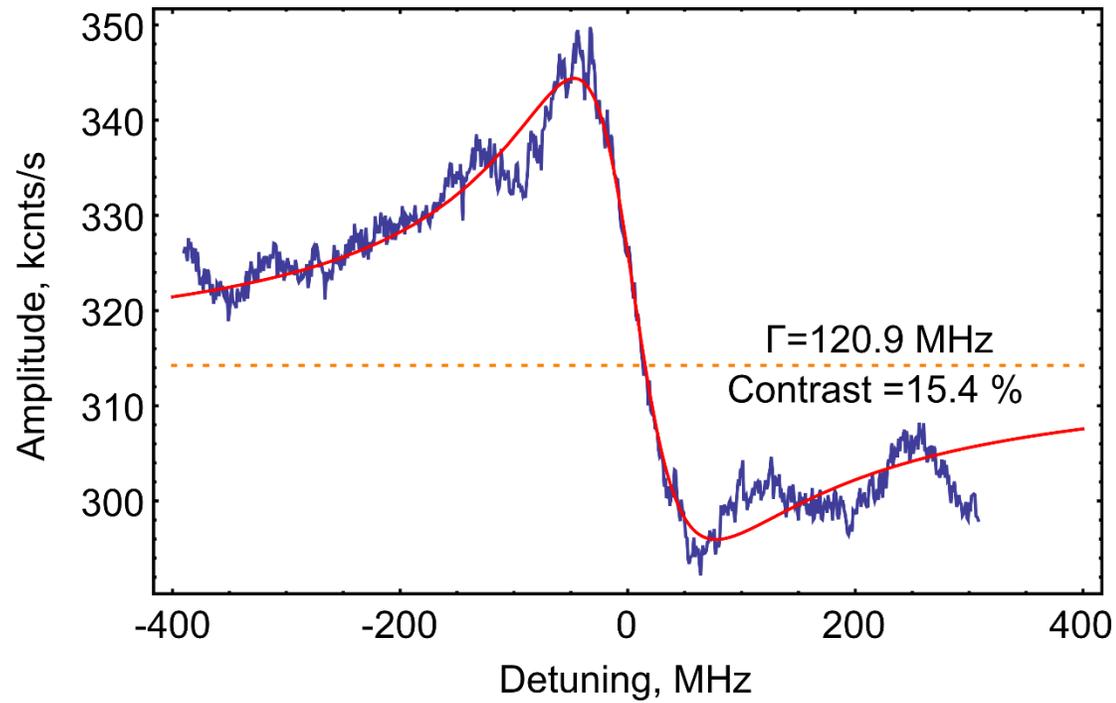
# Why so special?



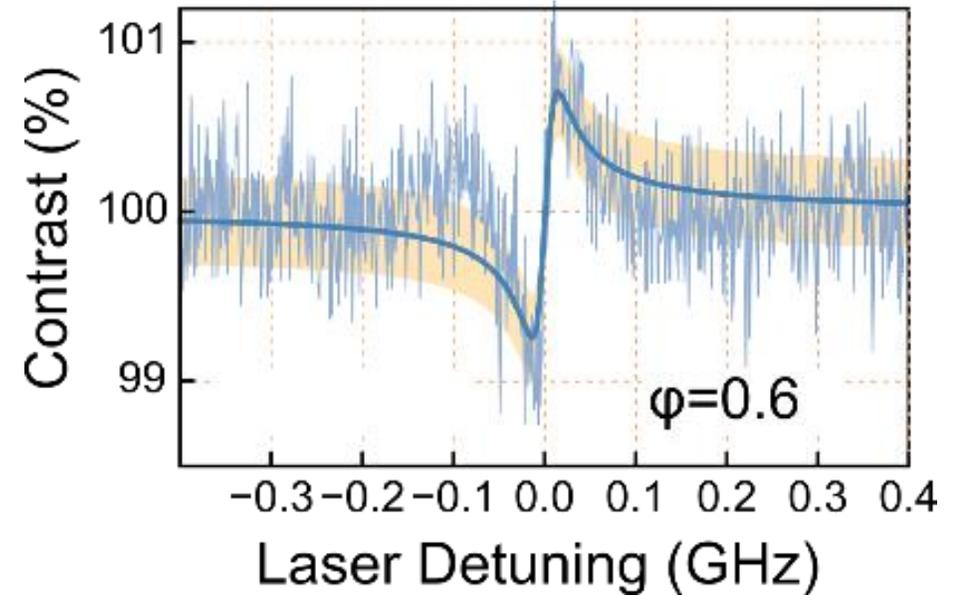
Filter is removed from the detection channel  
“extinction” measurements



# Why so special?

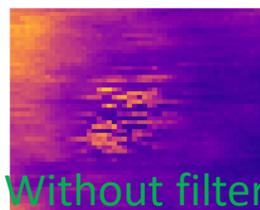
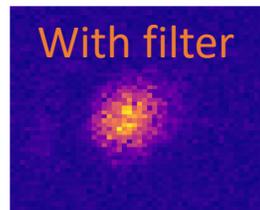
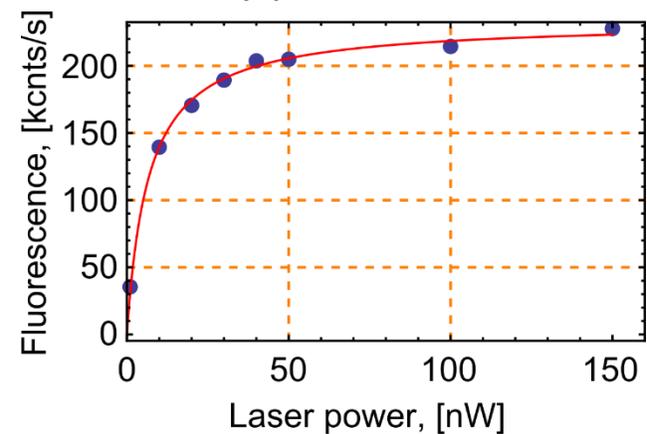


Extinction on NV center gives only about 2%

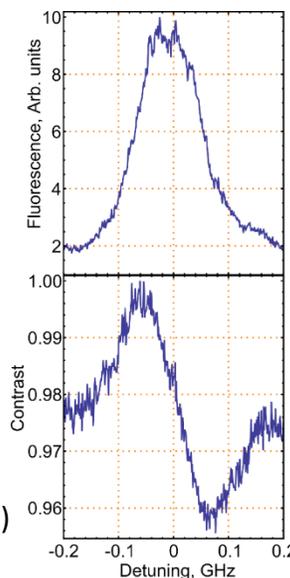
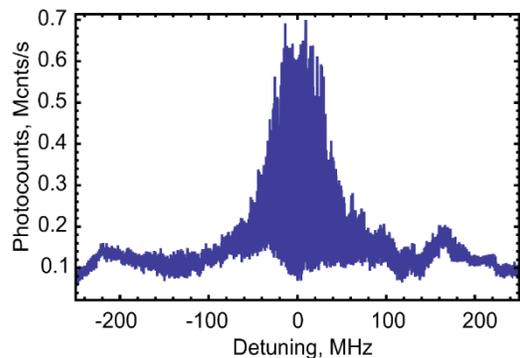


# Why so special?

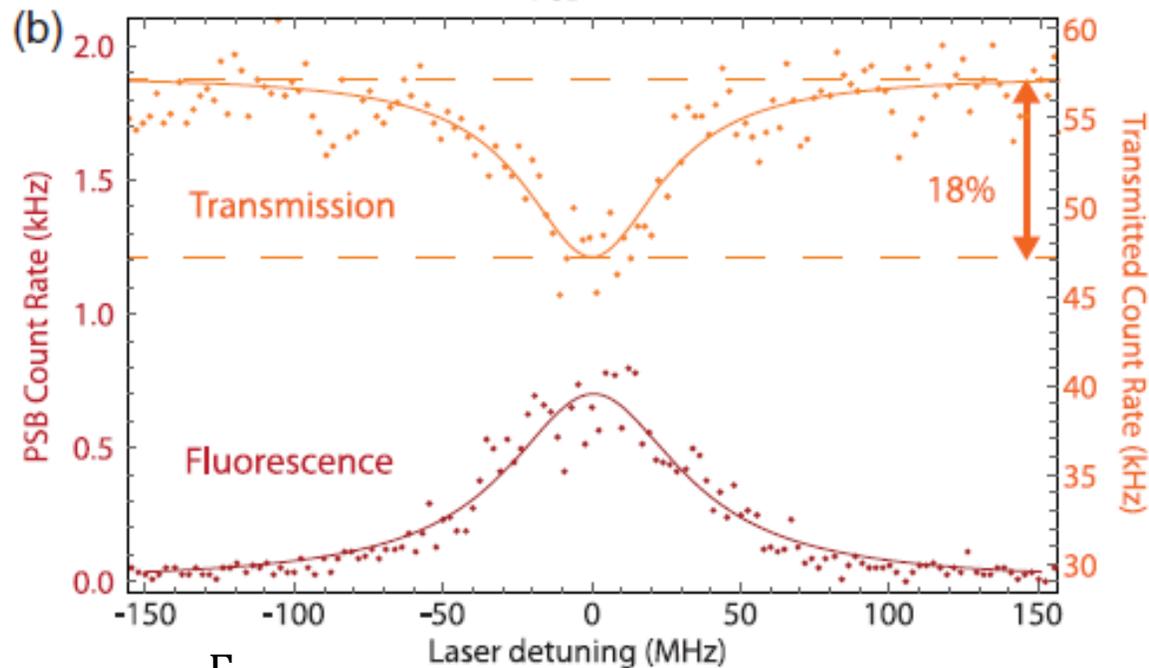
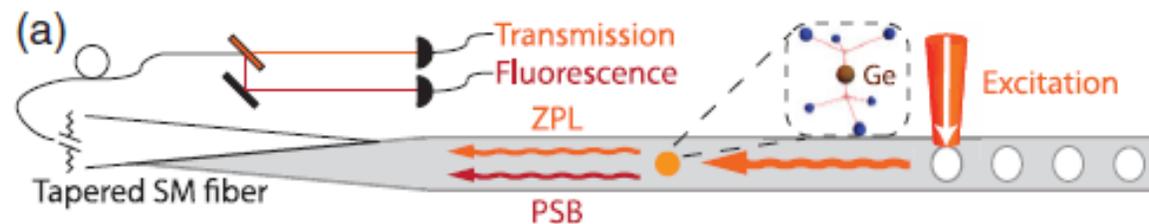
Only phonon sideband



All fluorescence



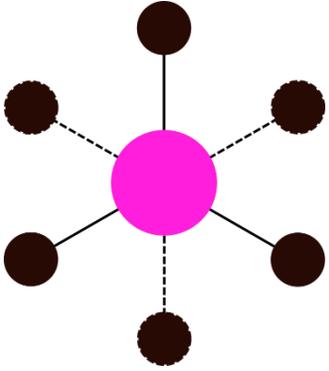
P. Siyushev et al. *Phys. Rev. B* **96**, 081201 (2017)



$$C = \frac{\Gamma_{1D}}{\Gamma'} \geq 0.1$$

M. K. Bhaskar et al. *Phys. Rev. Lett.* **118**, 223603 (2017)

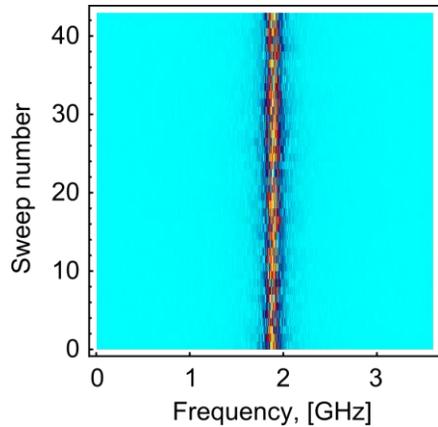
# Why so special?



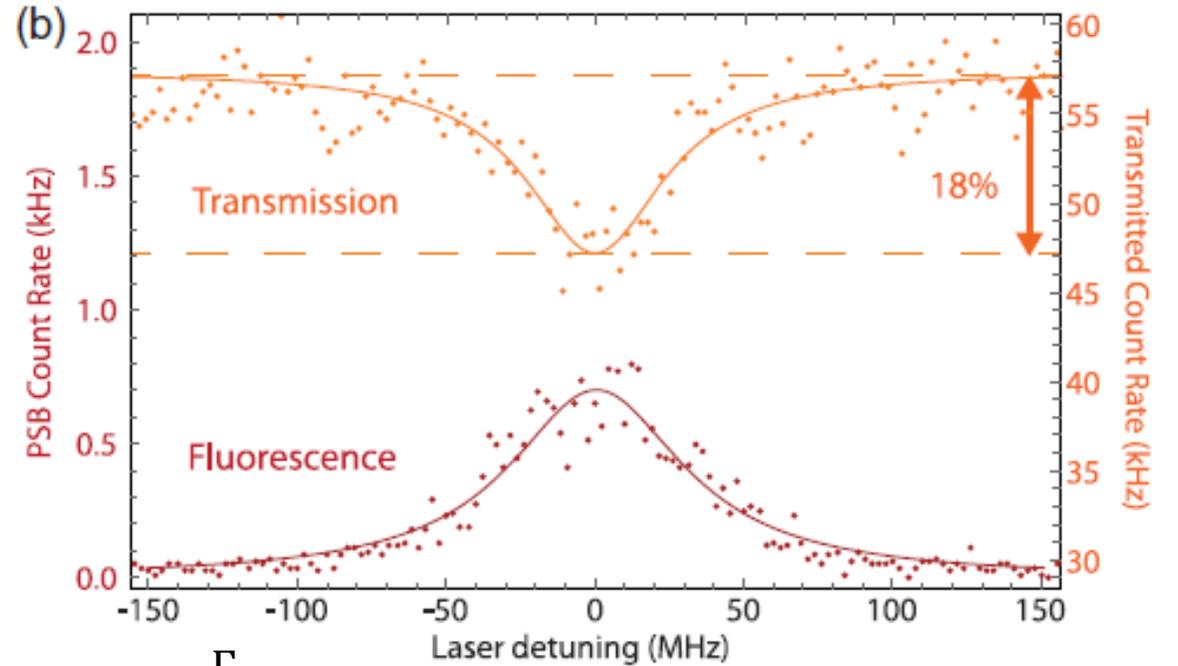
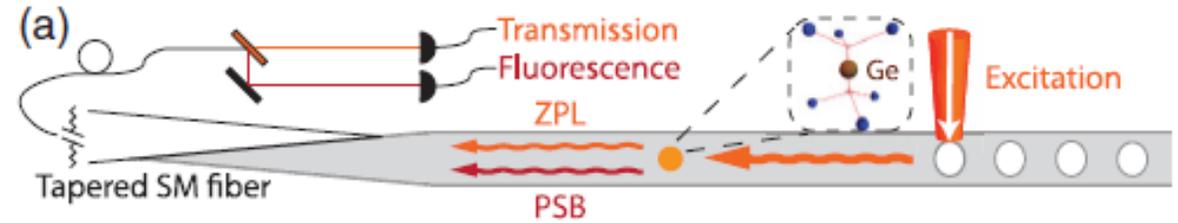
High Debye–Waller factor

$D_{3d}$  point group symmetry

~~$$\Delta E = -\mathbf{d}_{nm} \cdot \boldsymbol{\varepsilon} - \frac{1}{2} \alpha_{kl} \varepsilon_k \varepsilon_l$$~~



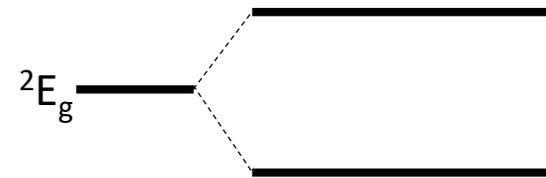
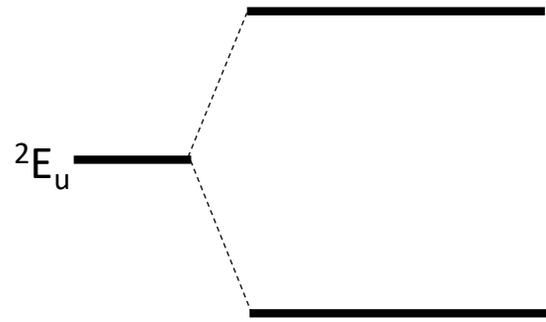
P. Siyushev et al. *Phys. Rev. B* **96**, 081201 (2017)



$$C = \frac{\Gamma_{1D}}{\Gamma'} \geq 0.1$$

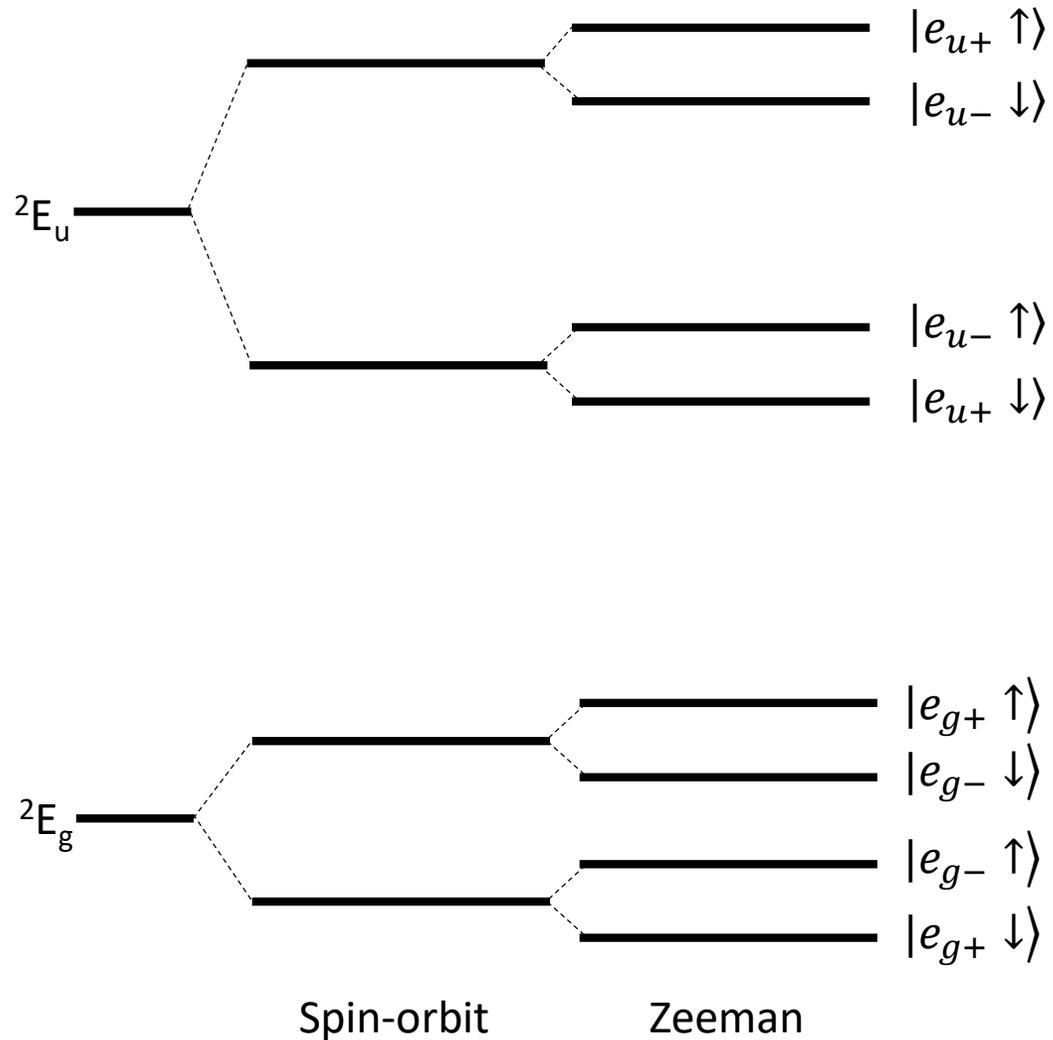
M. K. Bhaskar et al. *Phys. Rev. Lett.* **118**, 223603 (2017)

# Energy level structure



Spin-orbit

# Energy level structure



Ground state splitting:

~50 GHz (SiV),

~150 GHz (GeV),

~850 GHz (SnV),

~5700 GHz (PbV)

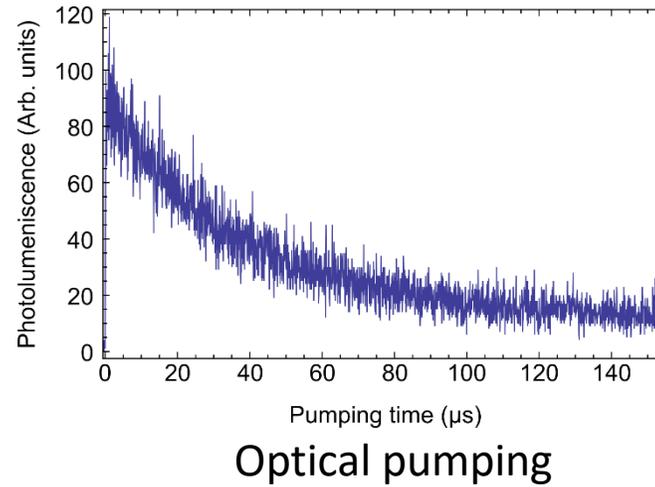
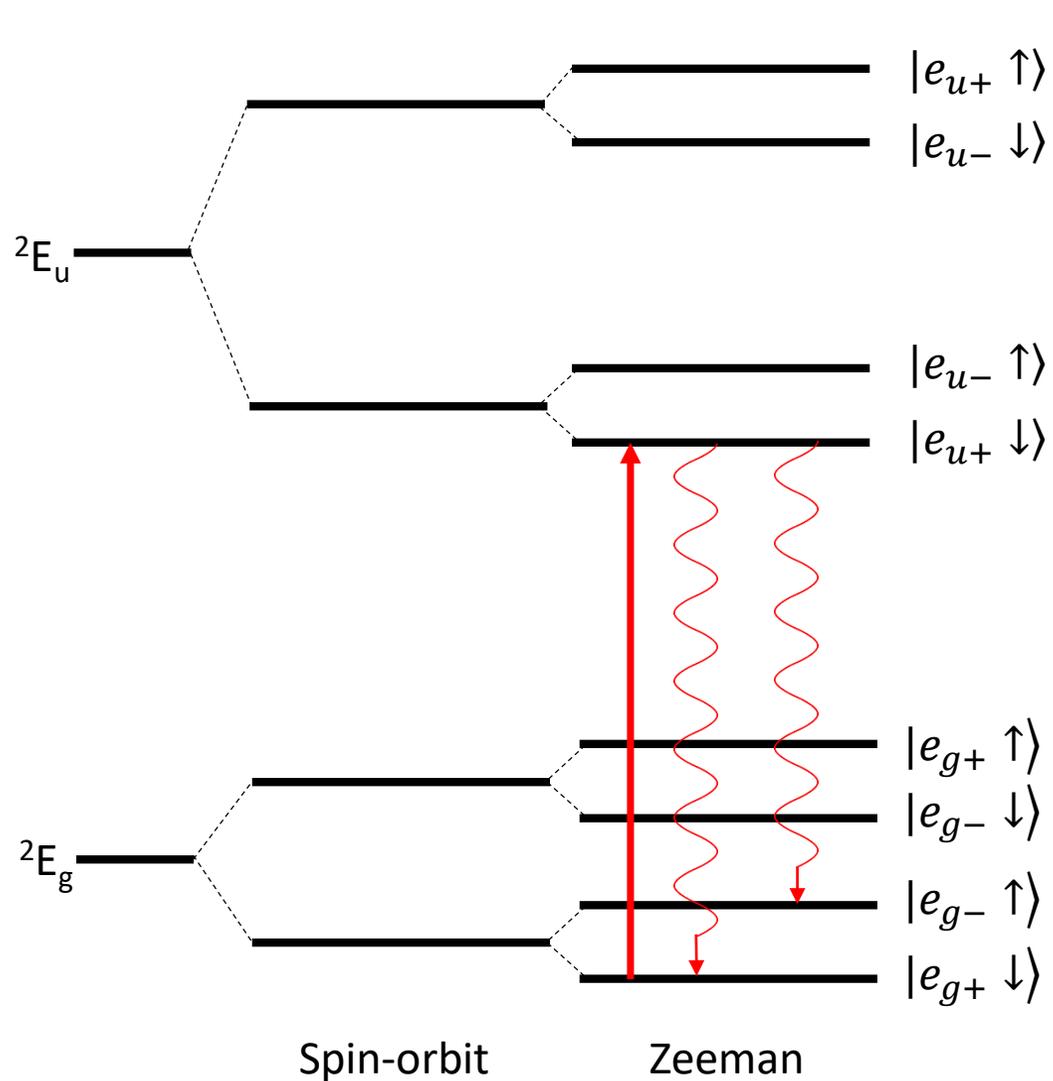
*C. Hepp et al. Phys. Rev. Lett. **112**, 036405 (2014)*

*P. Siyushev et al. Phys. Rev. B **96**, 081201 (2017)*

*T. Iwasaki et al. Phys. Rev. Lett. **119**, 253601 (2017)*

*M. Trusheim et. al, Phys. Rev. B **99**, 075430 (2019)*

# Energy level structure



Ground state splitting:  
 $\sim 50$  GHz (SiV),  
 $\sim 150$  GHz (GeV),  
 $\sim 850$  GHz (SnV),  
 $\sim 5700$  GHz (PbV)

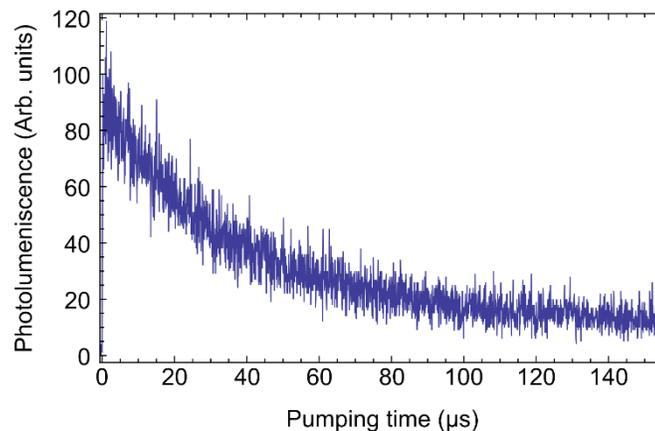
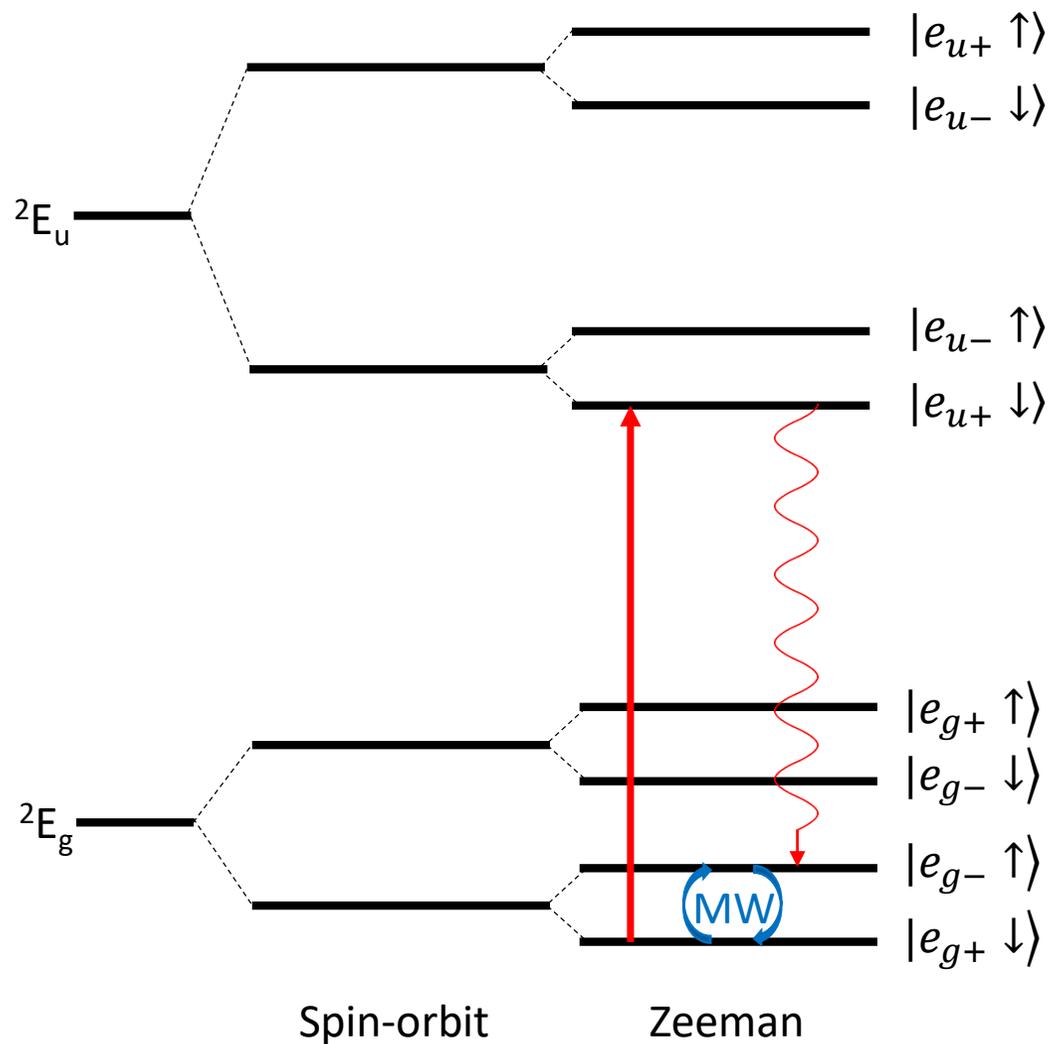
*C. Hepp et al. Phys. Rev. Lett. **112**, 036405 (2014)*

*P. Siyushev et al. Phys. Rev. B **96**, 081201 (2017)*

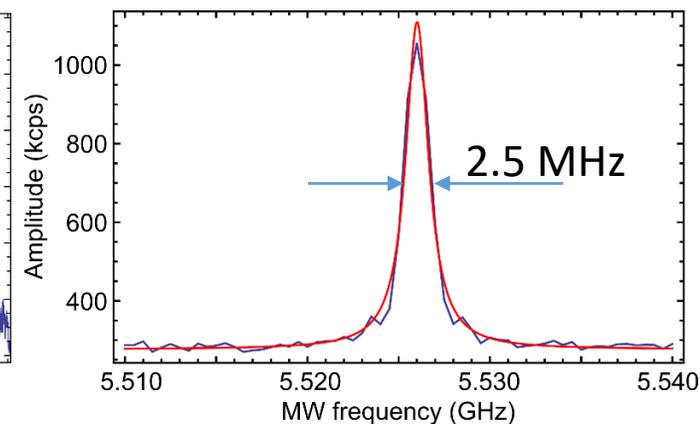
*T. Iwasaki et al. Phys. Rev. Lett. **119**, 253601 (2017)*

*M. Trusheim et. al, Phys. Rev. B **99**, 075430 (2019)*

# Energy level structure



Optical pumping



Example of ODMR

Ground state splitting:

- $\sim 50$  GHz (SiV),
- $\sim 150$  GHz (GeV),
- $\sim 850$  GHz (SnV),
- $\sim 5700$  GHz (PbV)

*C. Hepp et al. Phys. Rev. Lett. **112**, 036405 (2014)*

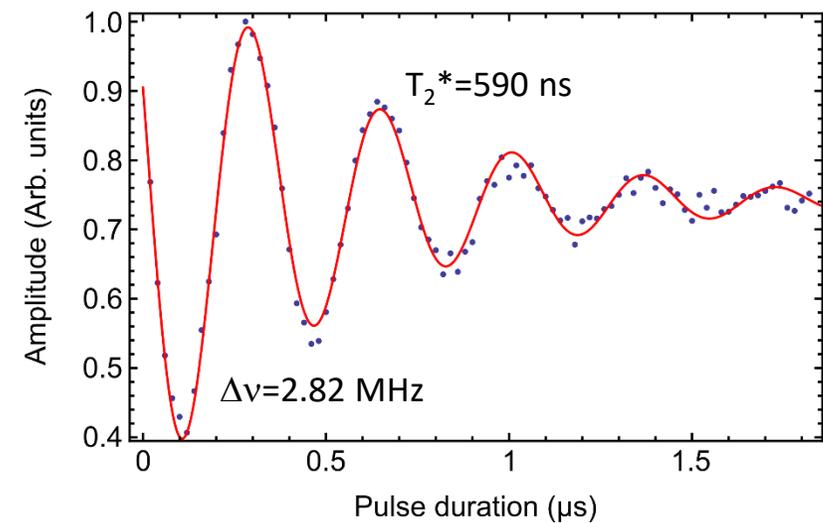
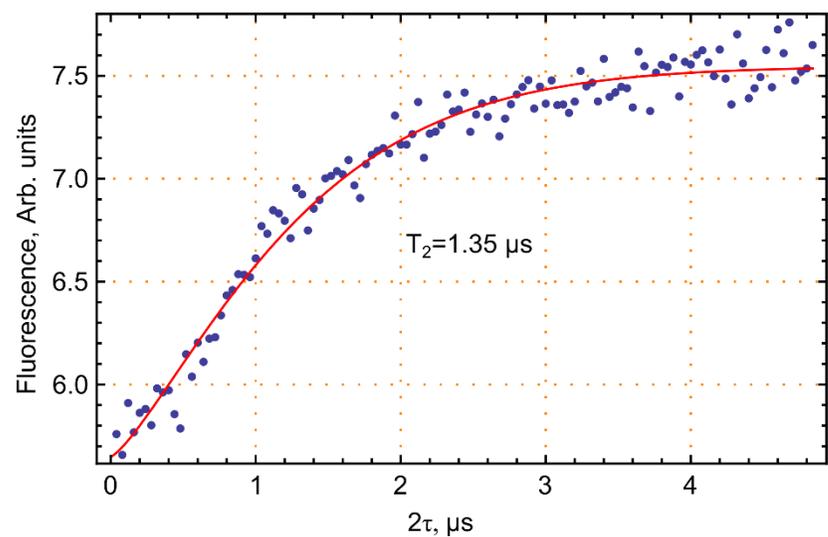
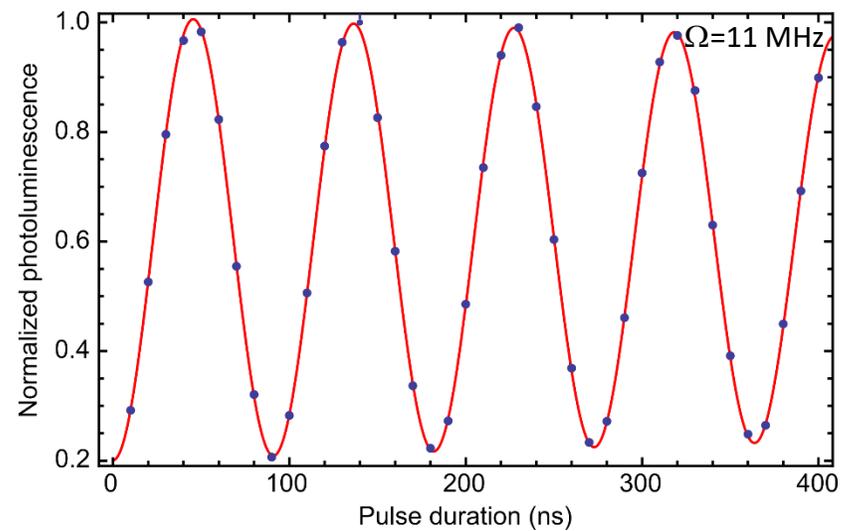
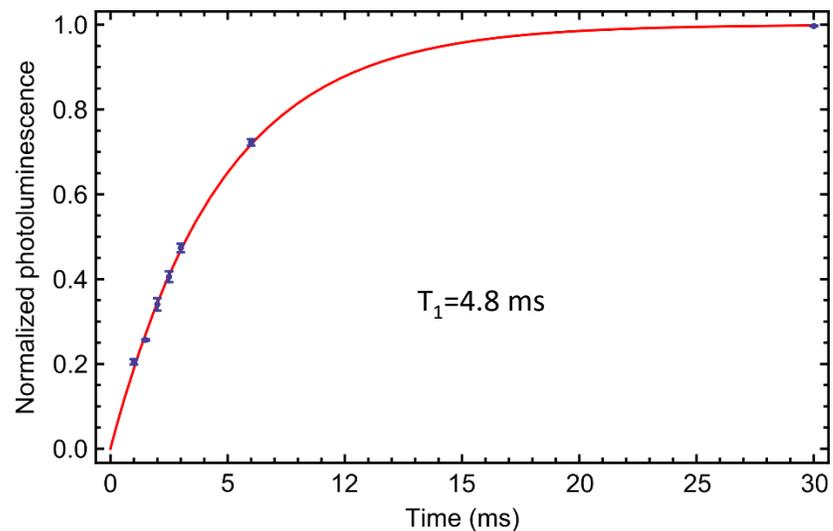
*P. Siyushev et al. Phys. Rev. B **96**, 081201 (2017)*

*T. Iwasaki et al. Phys. Rev. Lett. **119**, 253601 (2017)*

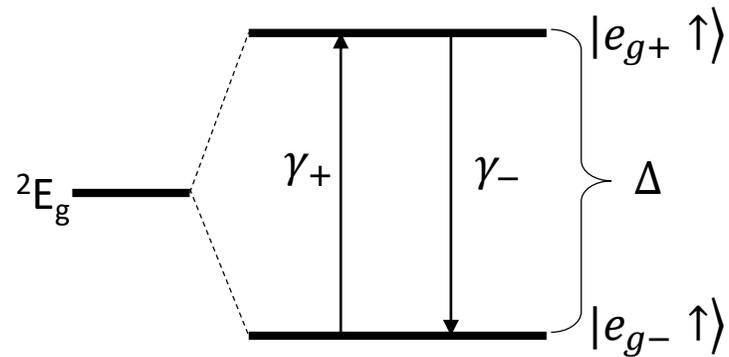
*M. Trusheim et. al, Phys. Rev. B **99**, 075430 (2019)*

# Electron spin properties of SiV center

$T \approx 2$  K



# Orbital relaxation



$$\gamma_+ = 2\pi\chi\rho\Delta^3 n(\Delta, T)$$

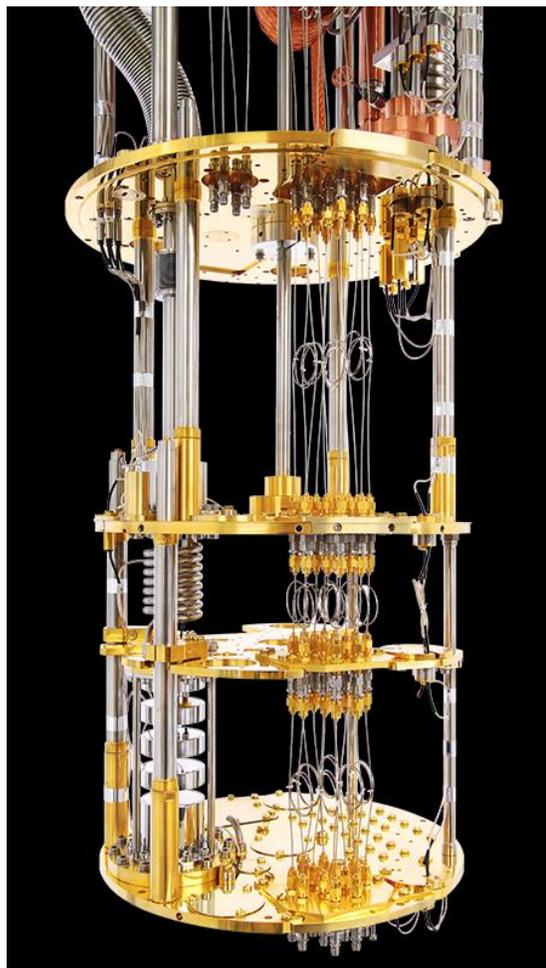
$$n(\Delta, T) = \frac{1}{e^{\frac{\hbar\Delta}{k_B T}} - 1}$$

$$\gamma_- = 2\pi\chi\rho\Delta^3 [n(\Delta, T) - 1]$$

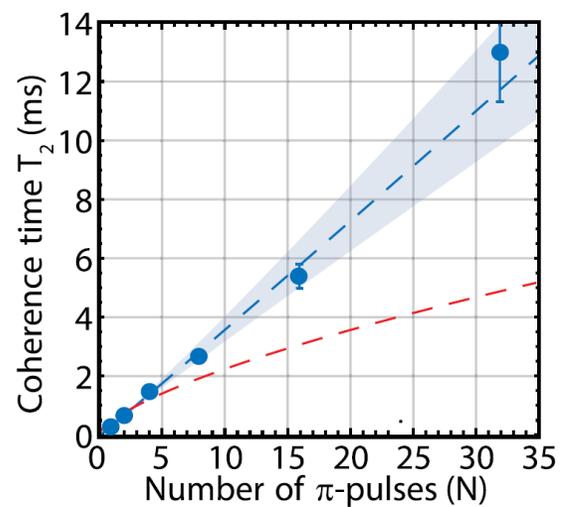
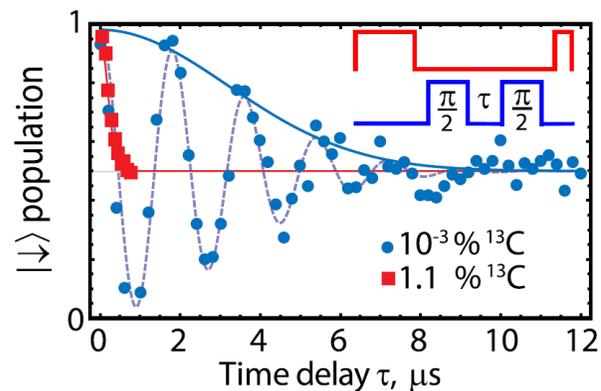
For  $T > \frac{\hbar\Delta}{k_B}$  linear scaling with temperature

in opposite case decreasing as  $e^{\frac{\hbar\Delta}{k_B T}}$

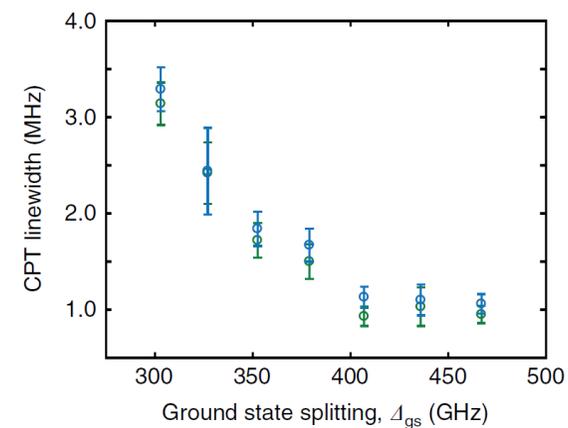
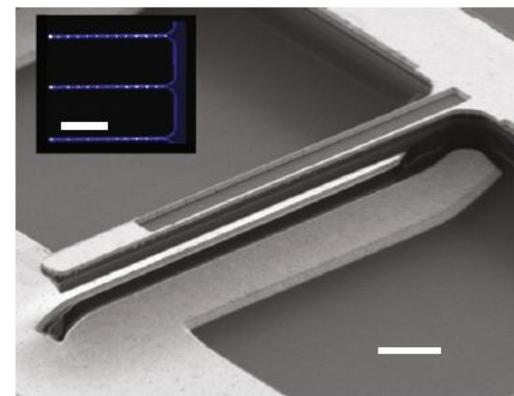
# How to avoid it?



D. Sukachev et. al, *Phys. Rev. Lett.* **119**, 223602 (2017)



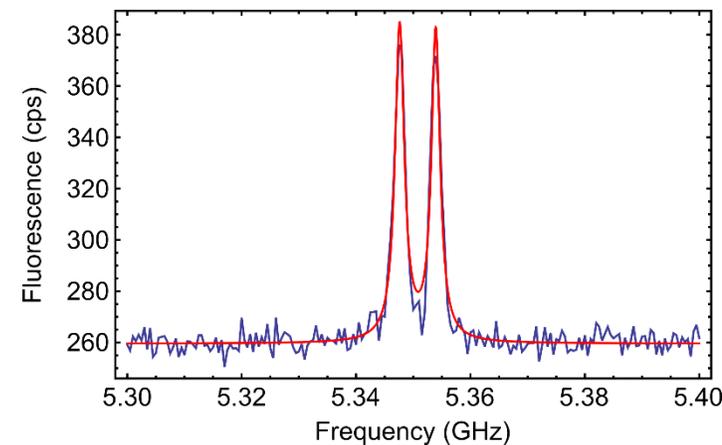
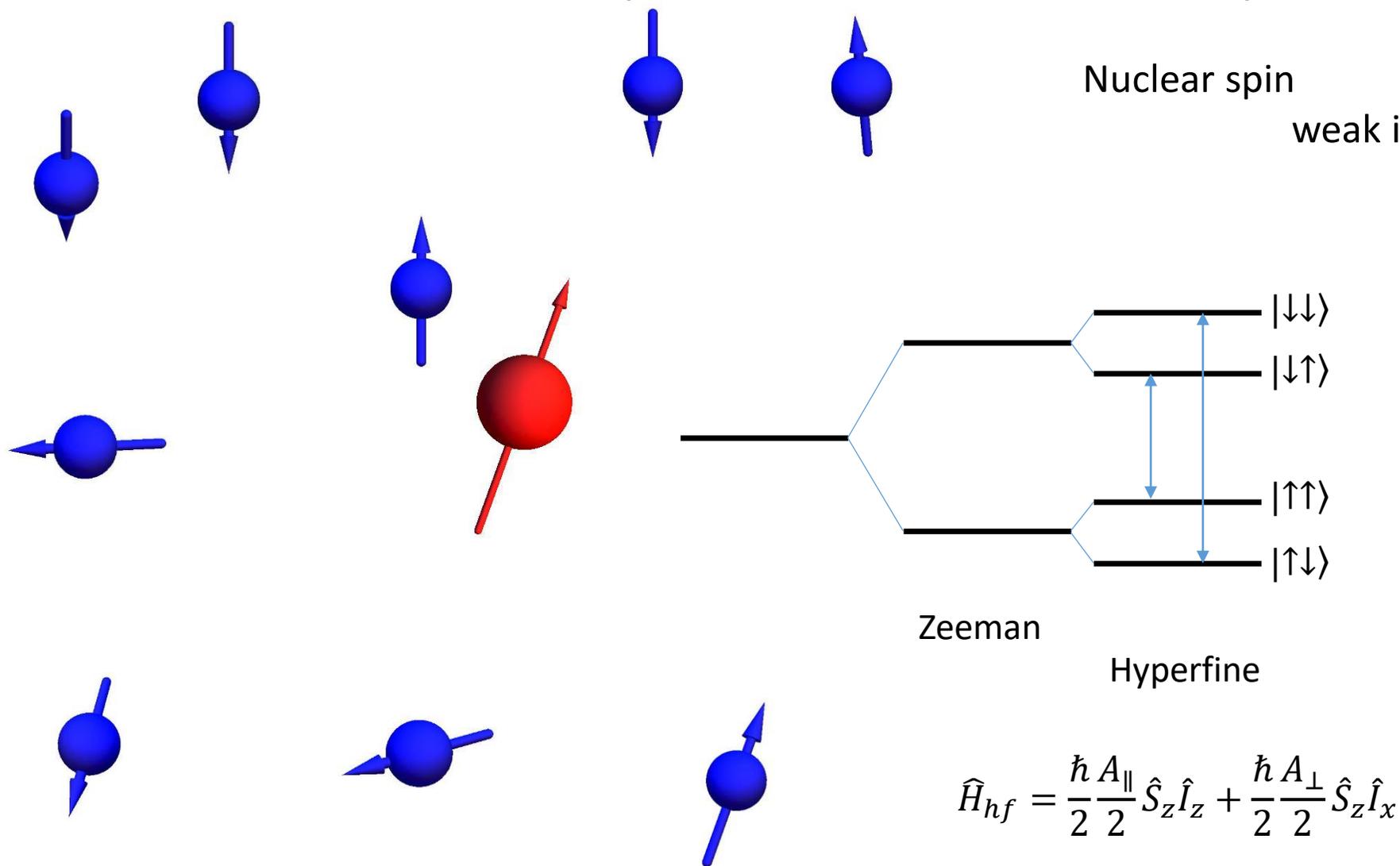
$T_2$  exceeds 10 ms @ 100 mK



$T_2^* = 0.25 \mu\text{s}$

Y.-I. Sohn et. al, *Nat. Commun.* **9**, 2012 (2018)

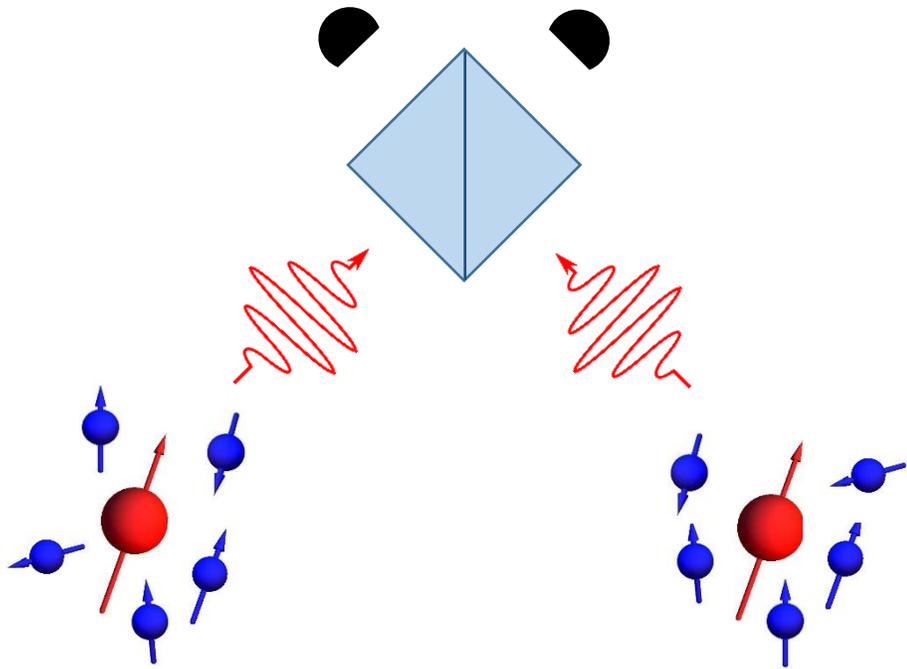
# Electron coupled to nuclear spins



Splitting  $\sim 6.3$  MHz

Linewidth  $\sim 2$  MHz

# Electron coupled to nuclear spins



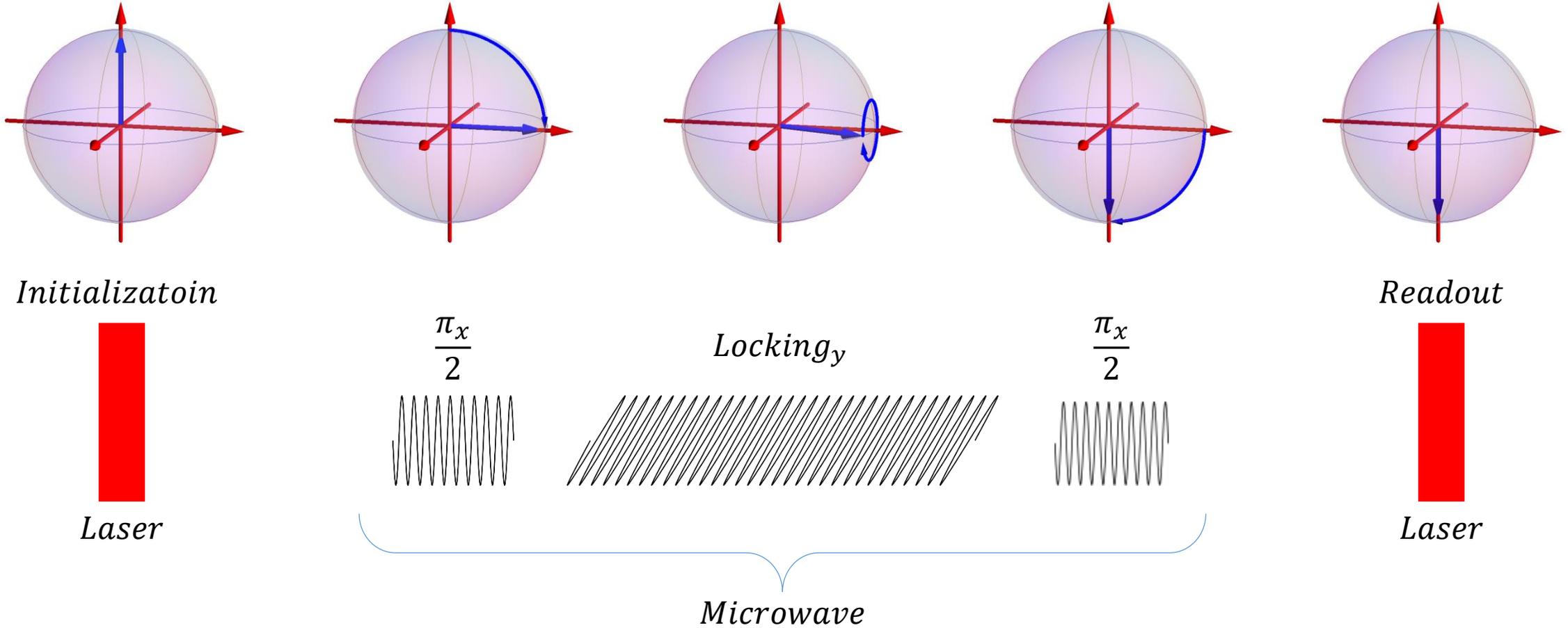
Overall strategy:

- Use Si(Ge,Sn...)V only as SPIN-PHOTON interface
- Use nuclear spin as a stationary qubit

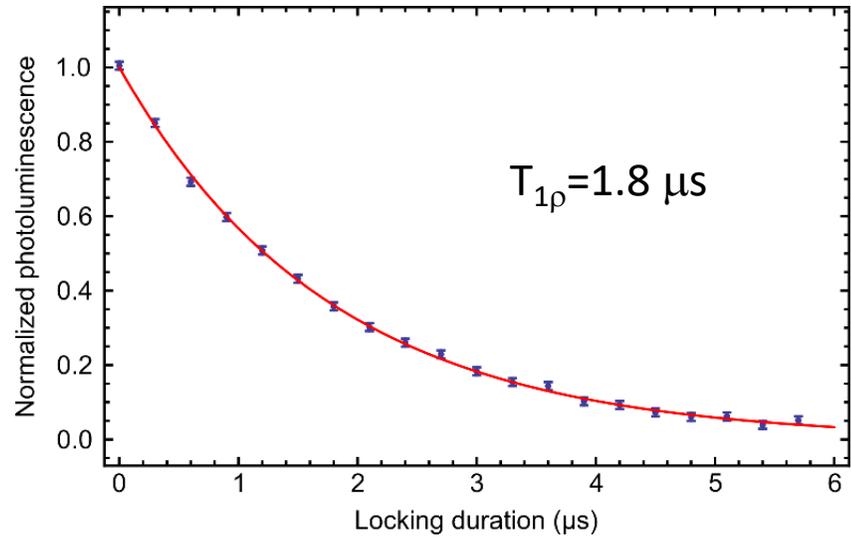
Nuclear spin should be

- initialized
- readout

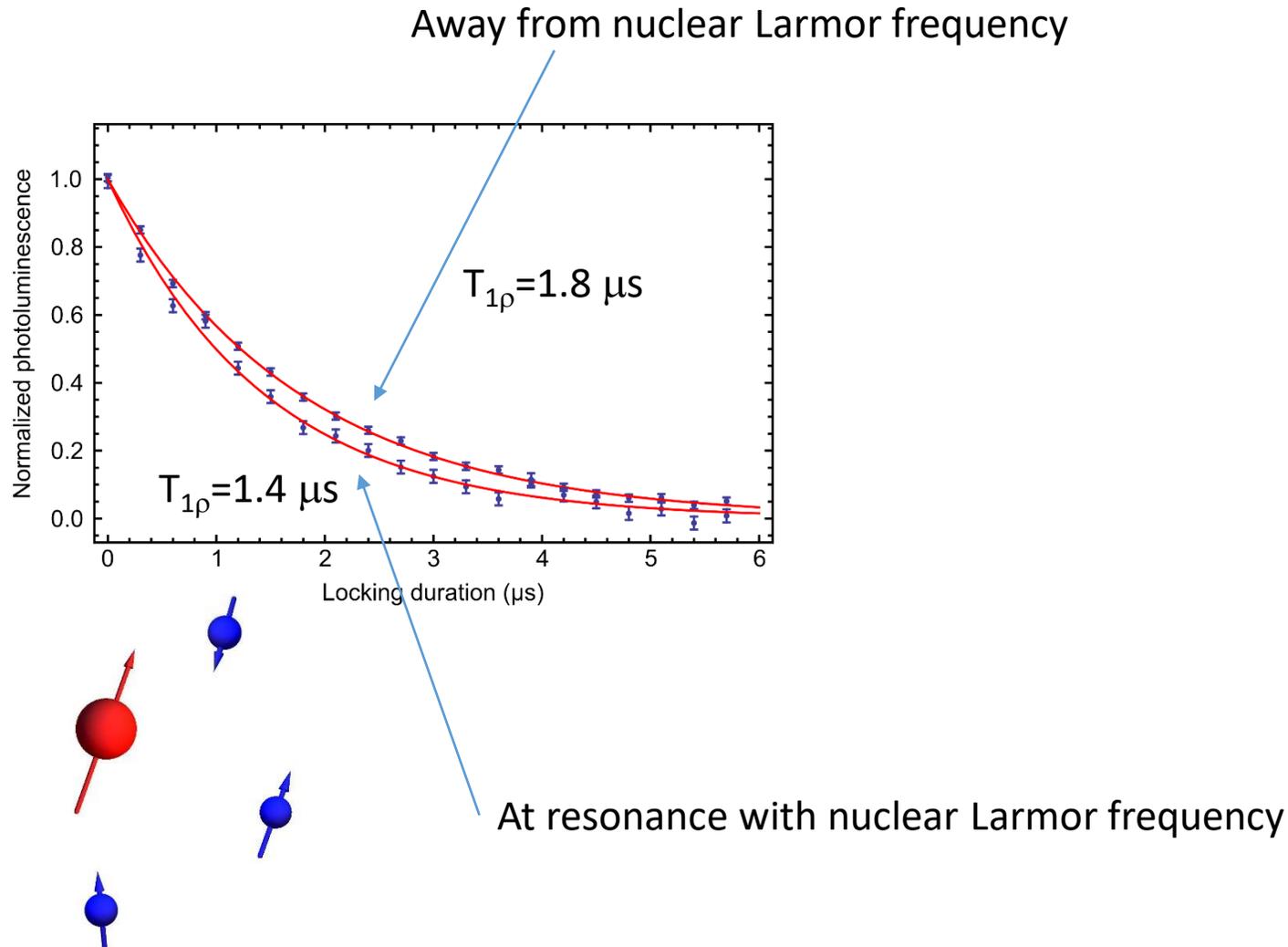
# Spin locking



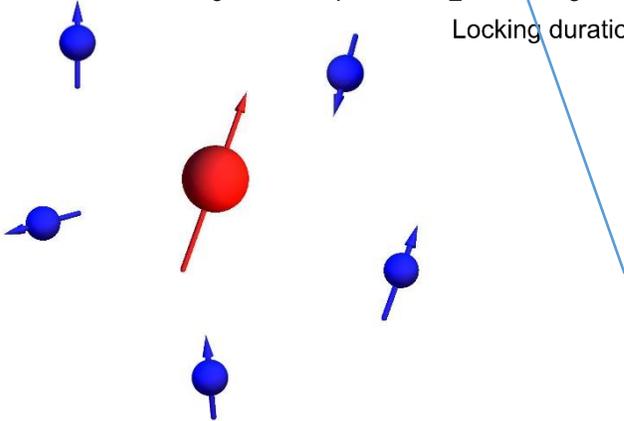
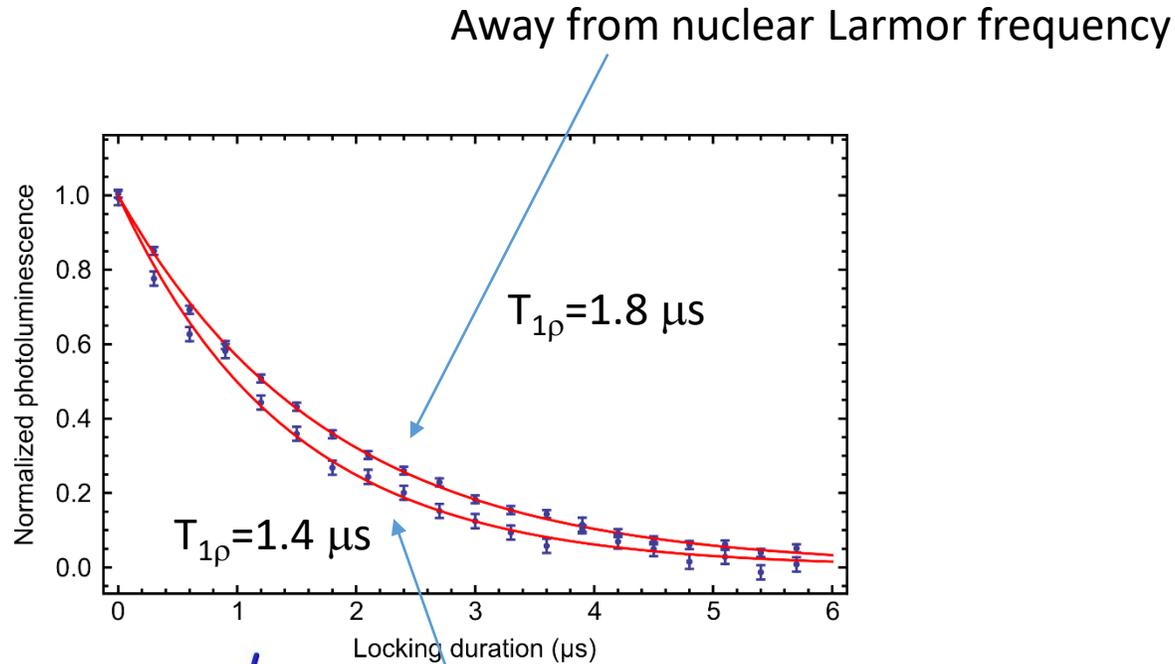
# Spin locking and Hartmann-Hahn



# Spin locking and Hartmann-Hahn

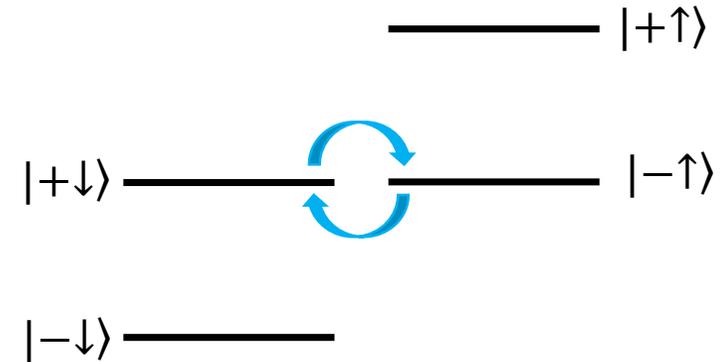


# Spin locking and Hartmann-Hahn

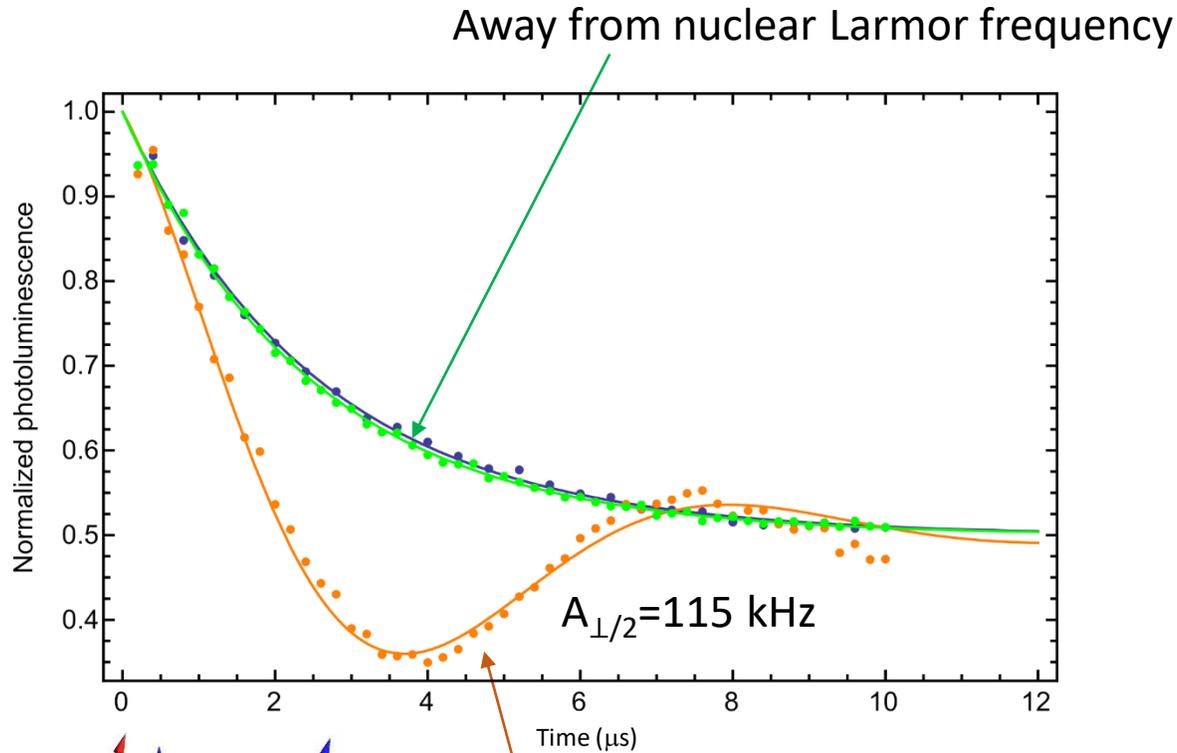


At resonance with nuclear Larmor frequency

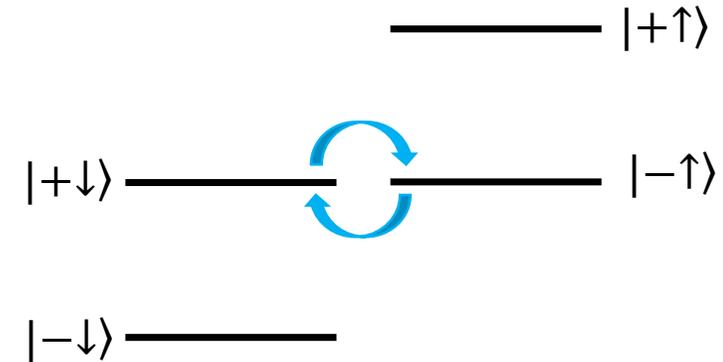
$$\Omega = \omega_L = \gamma_N B / \hbar \quad \text{Hartmann-Hahn condition}$$



# Spin locking and Hartmann-Hahn



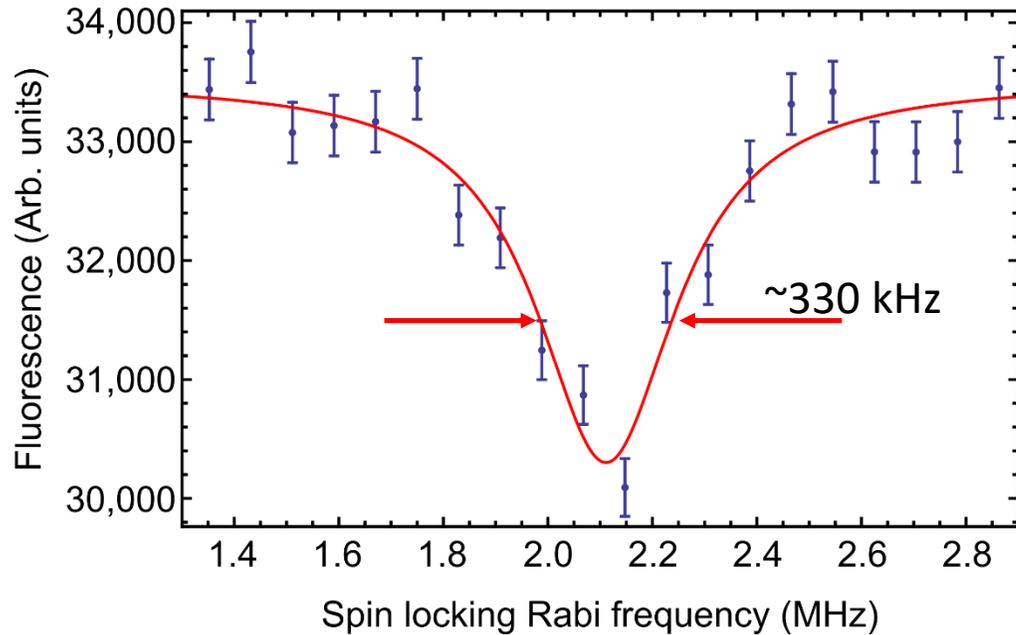
$$\Omega = \omega_L = \gamma_N B / \hbar \quad \text{Hartmann-Hahn condition}$$



At resonance with nuclear Larmor frequency

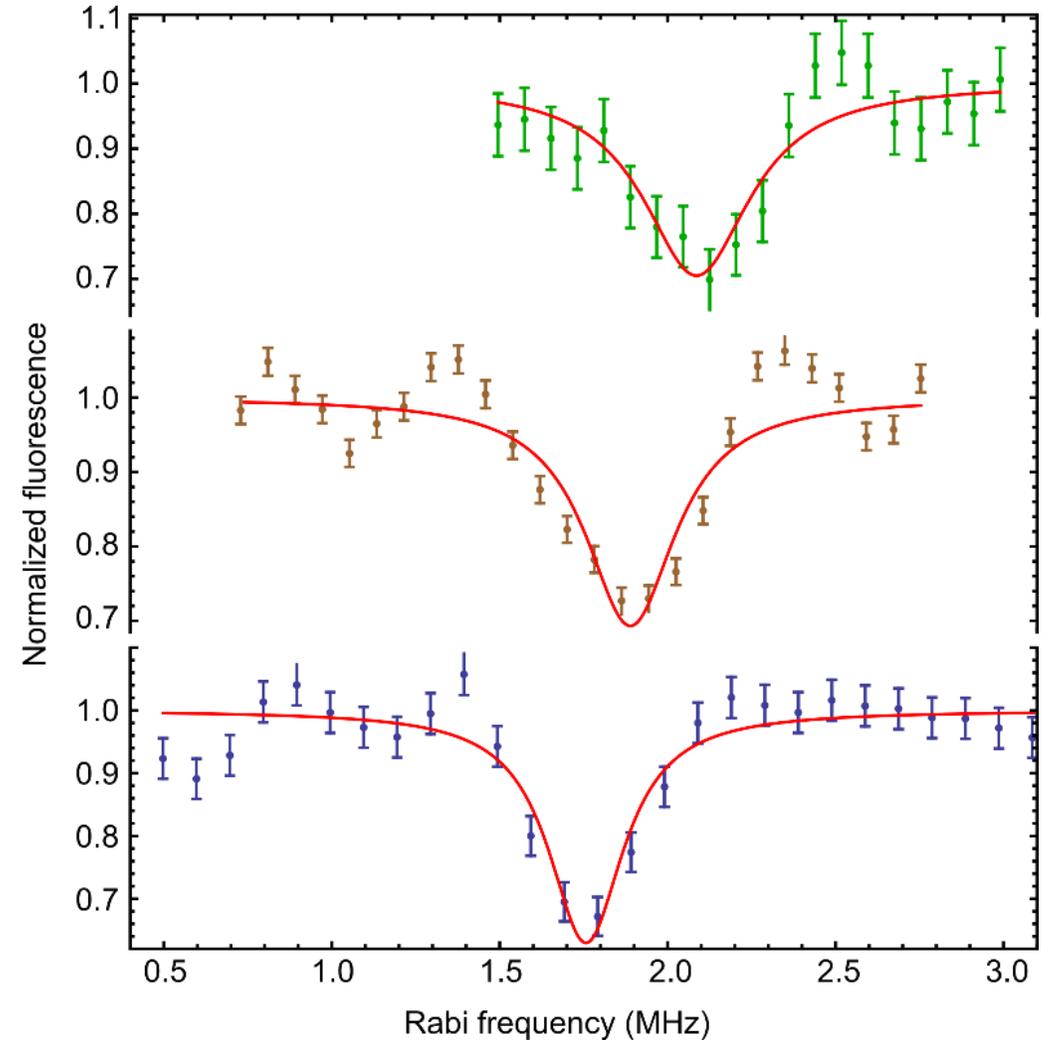
Oscillatory behavior reveals coupling to a single nuclear spin

# Nuclear Larmor frequency

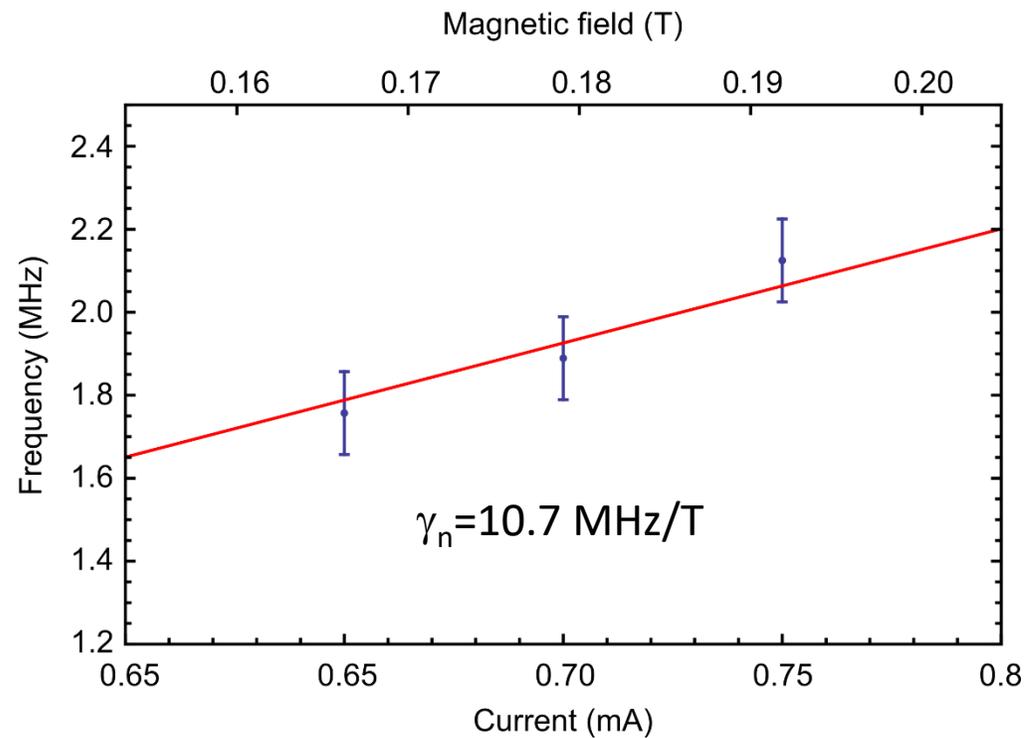
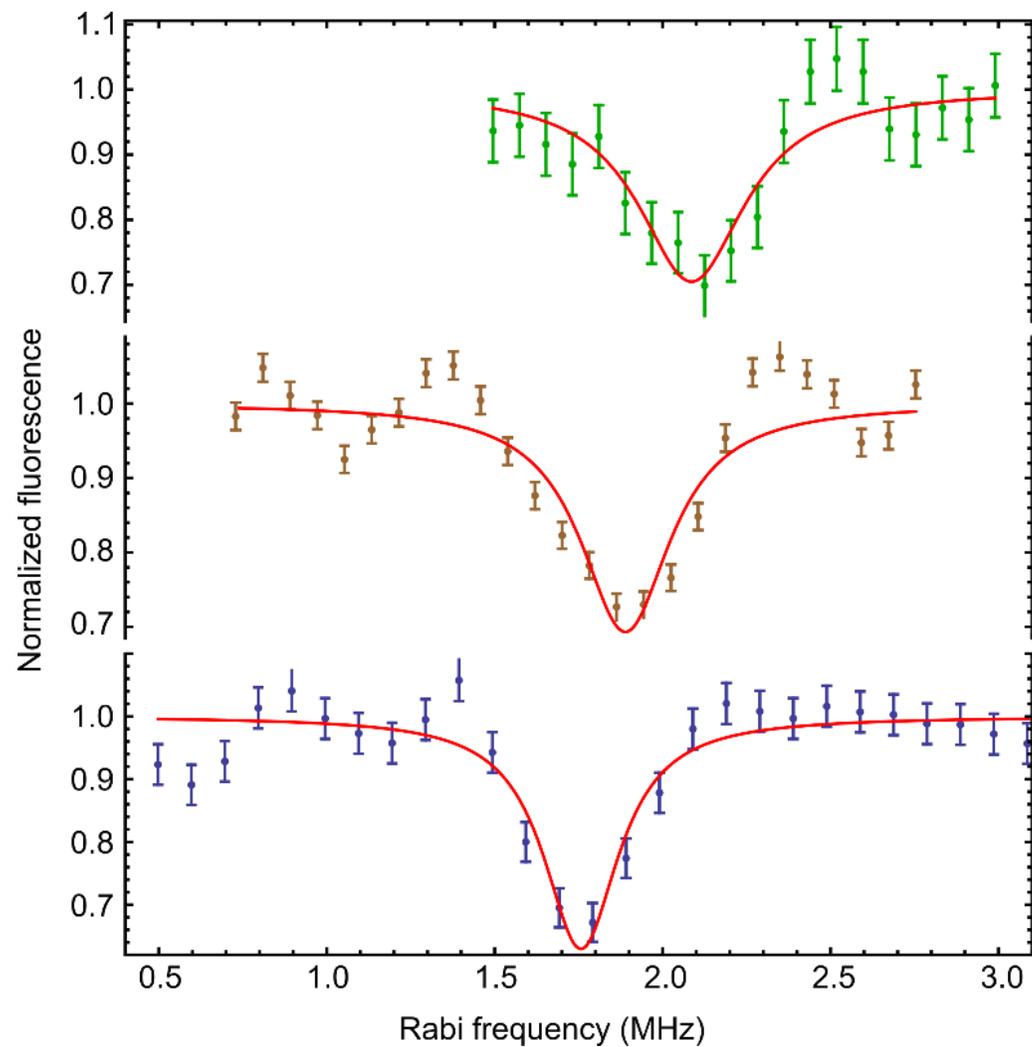


Larmor frequency of 2.1 MHz  
corresponds to 1970 G.

Width of the dip is most probably governed by the short pulse  
length of the locking pulse (in the order of  $1 \mu\text{s}$ )

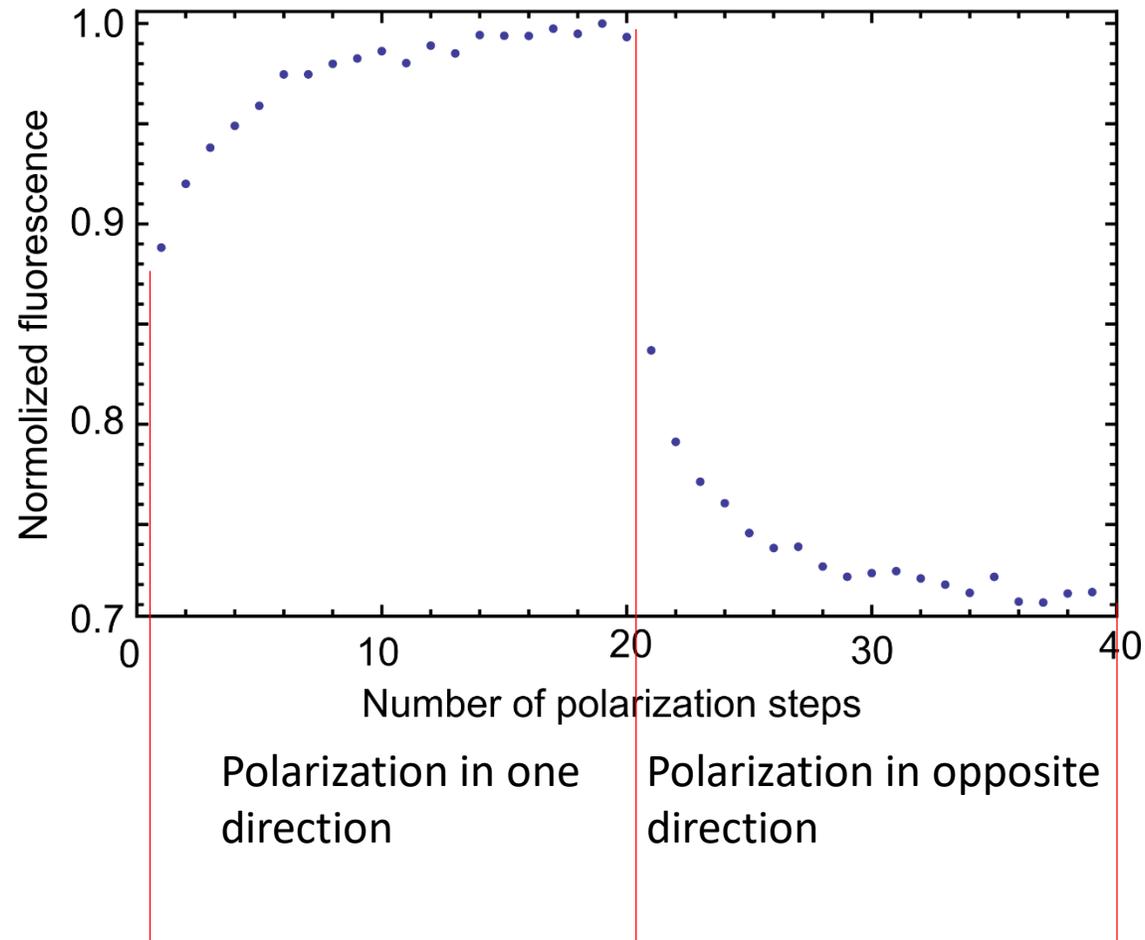


# Nuclear Larmor frequency



Carbon-13 gyromagnetic ratio  $\gamma_n = 10.705 \text{ MHz/T}$

# Polarization of $^{13}\text{C}$ spin bath



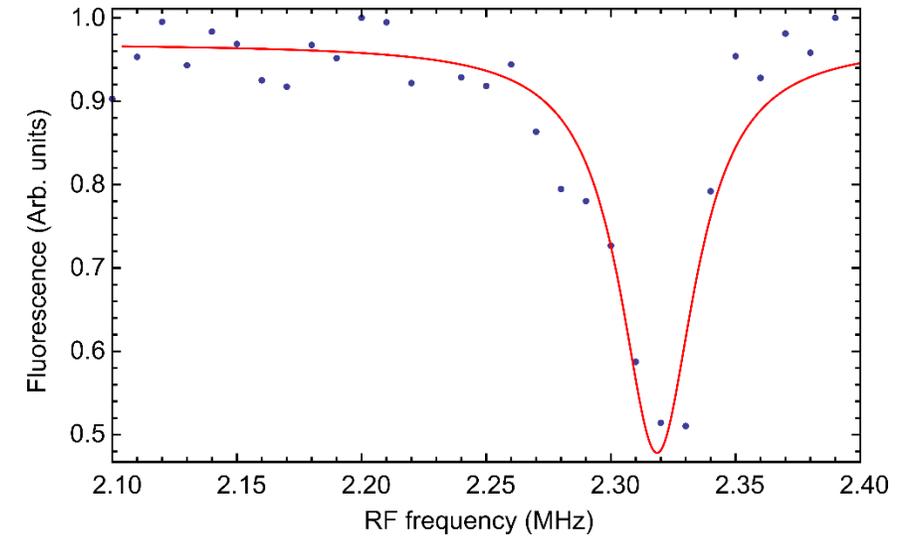
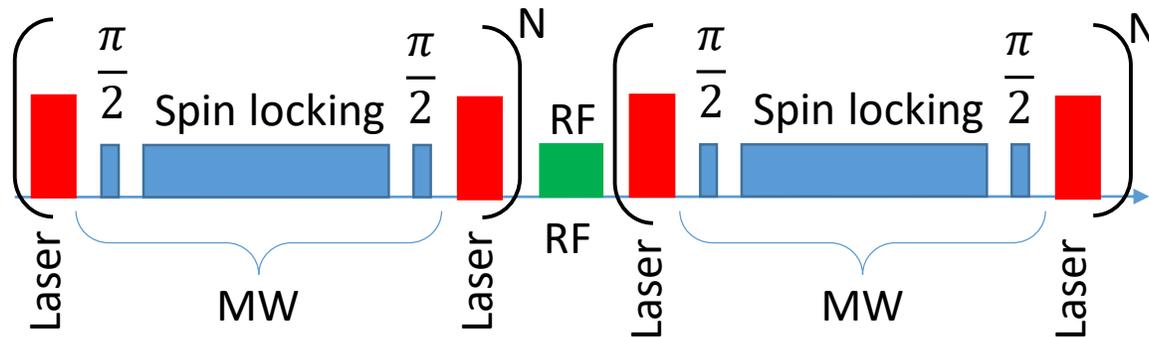
Polarization of a spin bath using Hartmann-Hahn technique

This can be used to readout nuclear spin state

# Addressing individual nuclear spin

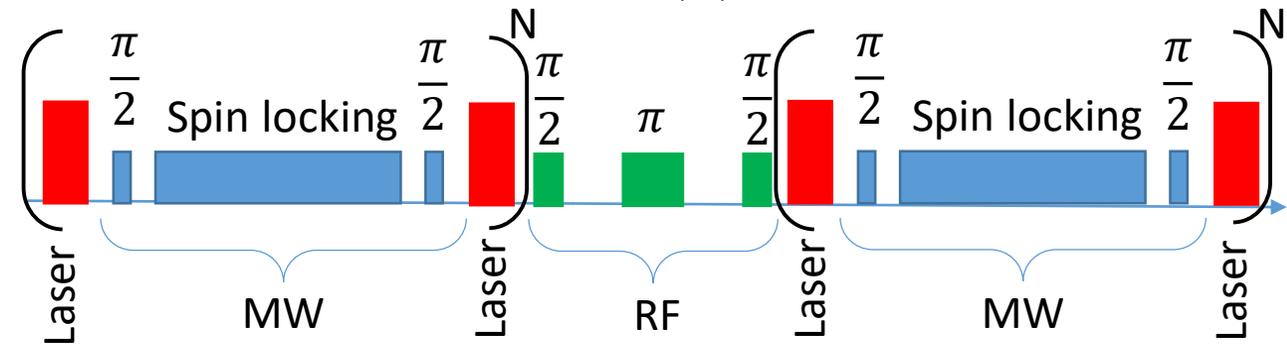
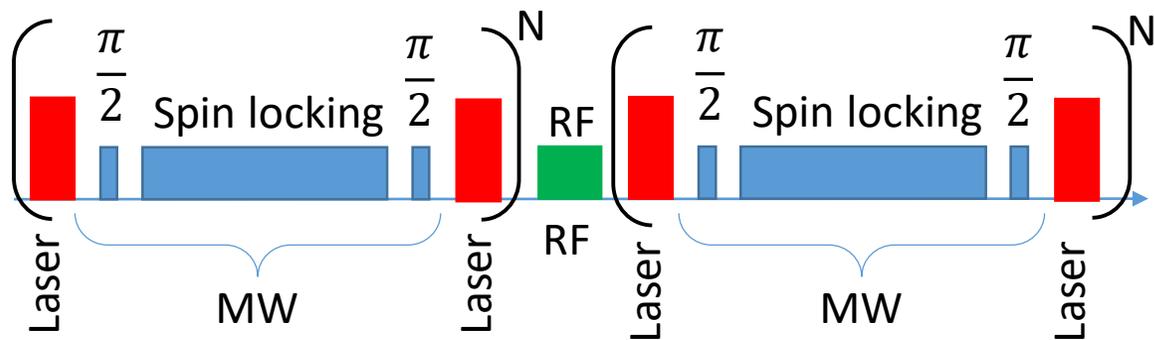
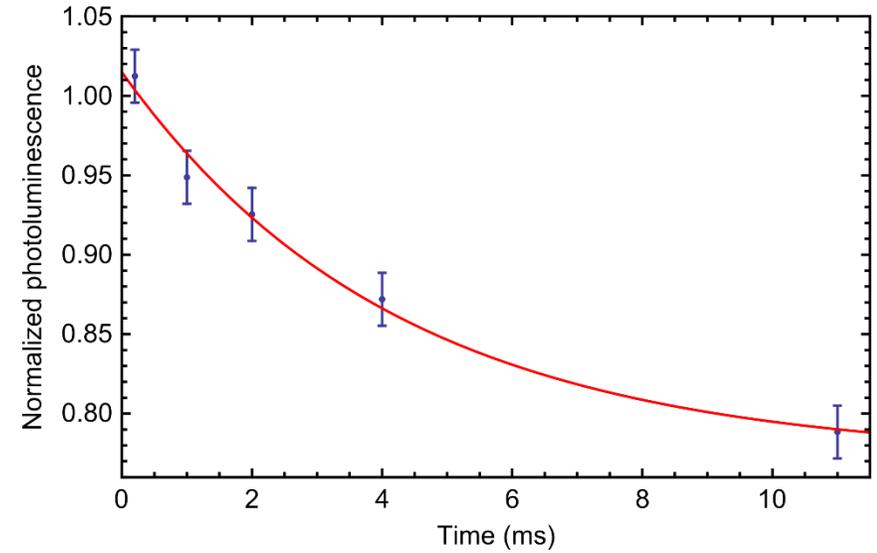
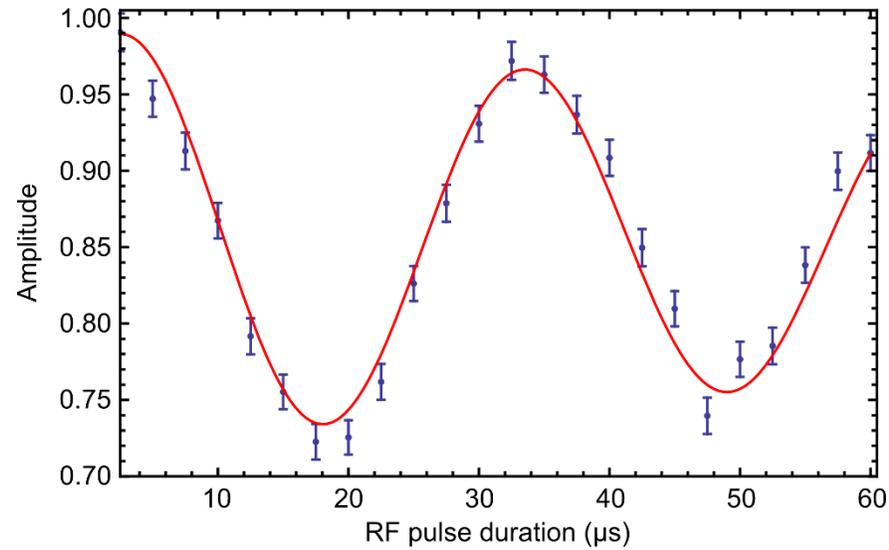
The nuclear resonance is shifted from the Larmor by  $A_{||}/2$

RF frequency is swept

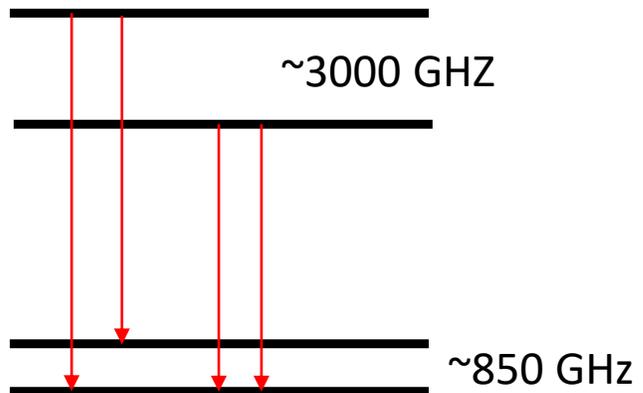
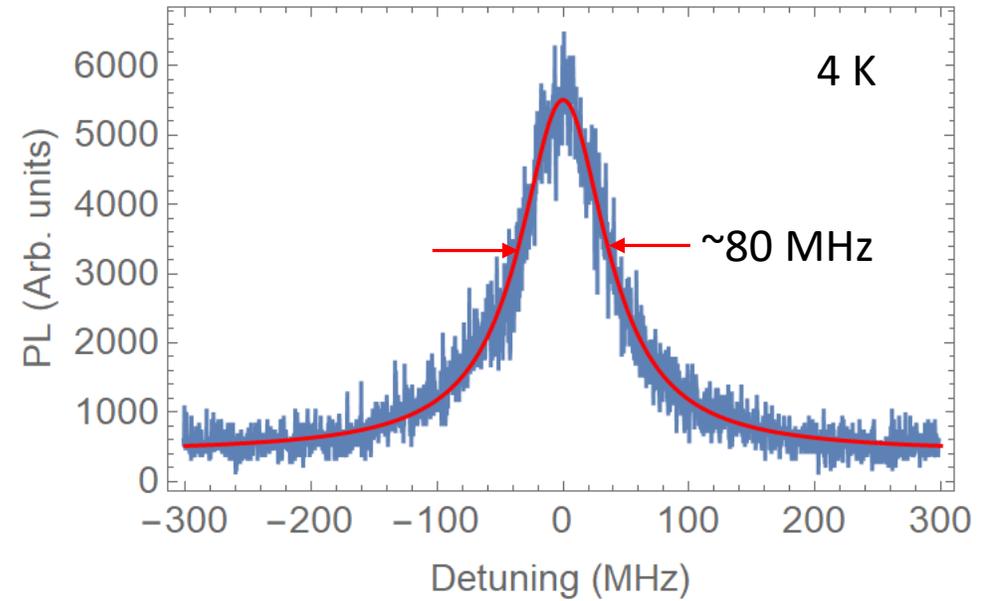
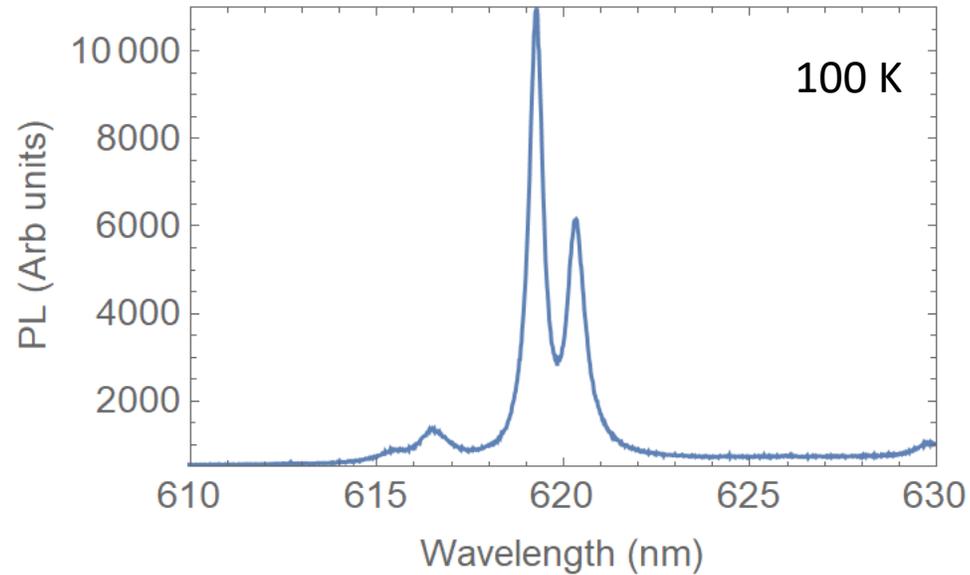


$$\begin{aligned}\omega_L &= 2\pi \times 1.96 \text{ MHz} \\ A_{||}/2 &= 360 \text{ KHz} \\ \omega_{\text{res}} &= 2\pi \times 2.32 \text{ MHz}\end{aligned}$$

# Readout of nuclear spins



# SnV spectra

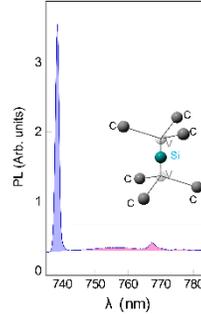


Not lifetime limited ( $\sim 30$  MHz)...

The relaxation time should increase by factor of 17

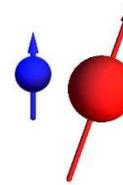
# Conclusion

- Excellent optical properties



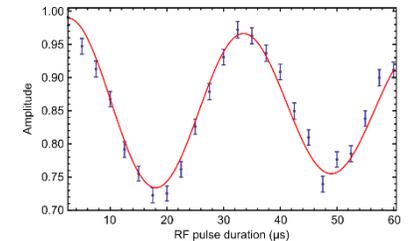
- Electron spin can be initialized, coherently manipulated and read out

- Defect can be coupled to nuclear spin



- Nuclear spin can be polarized and read out via electron spin

- SnV should enable longer electron coherence time



# Our collaborators who produce diamonds

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Igor N. Kupriyanov

Yuri M. Borzdov

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Sobolev Institute of Geology and Mineralogy)*

Takayuki Iwasaki

*(Tokyo Institute of Technology)*

Jocelyn Achard

Alexandre Tallaire

*(Université Paris 13)*

# Acknowledgements



Thank you for your attention!