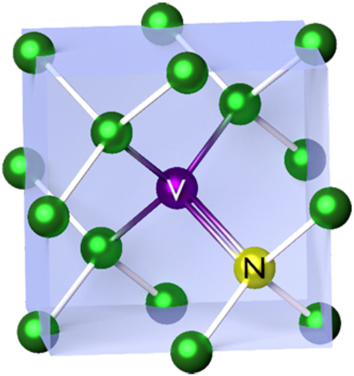
Group meeting : Cross-relaxation with NV centers ensemble in diamond

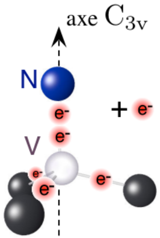
October 11, 2021

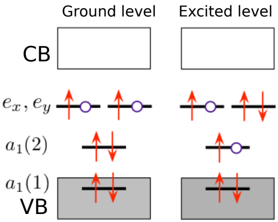
The Nitrogen Vacancy Center

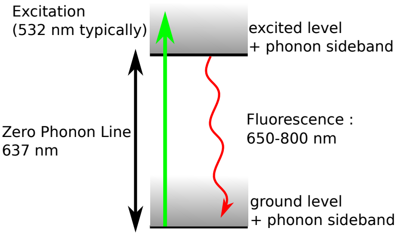


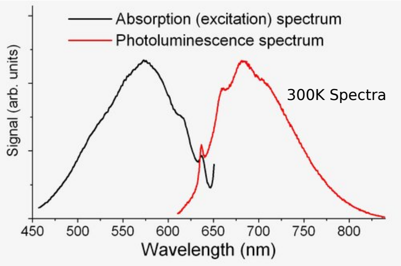
- 0D fluorescent object with ZPL at 638 nm
- Controllable and readable spin at room temperature (!)
- Working with 10^9 emitters (typ.)

NV center : Optical properties

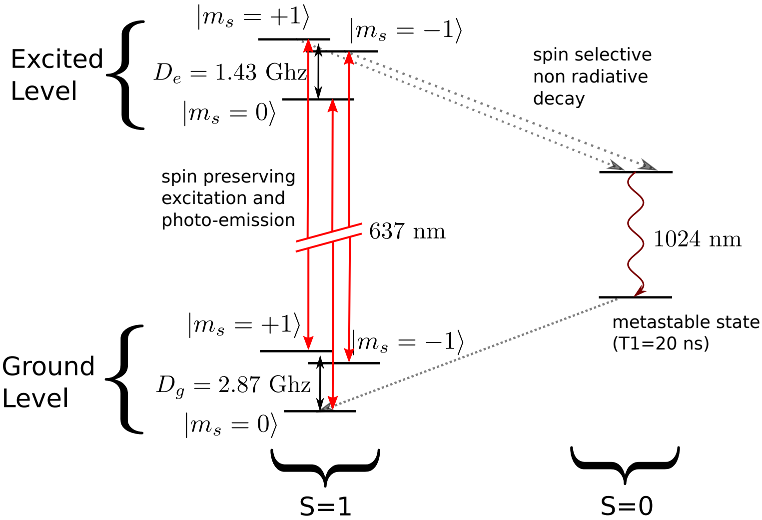




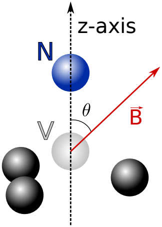




NV center : 8 levels



NV center : 3 levels

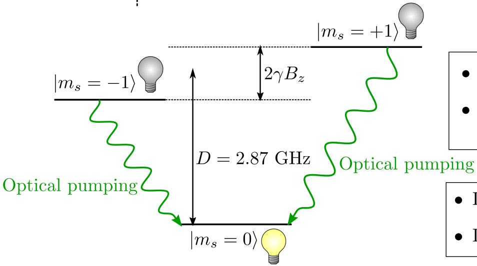


Ground level spin Hamiltonian

$$\hat{\mathcal{H}}_s = DS_z^2 + \gamma_e \mathbf{B} \cdot \hat{\mathbf{S}}$$

$D = 2.87\,\mathrm{GHz}$ and $\gamma_e = 2.8\,\mathrm{MHz/G}$

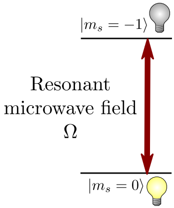
$$\mathcal{H}_s = \begin{pmatrix} D - \gamma_e B\cos\theta & \gamma_e B\sin\theta & 0 \\ \gamma_e B\sin\theta & 0 & \gamma_e B\sin\theta \\ 0 & \gamma_e B\sin\theta & D + \gamma_e B\cos\theta \end{pmatrix}$$

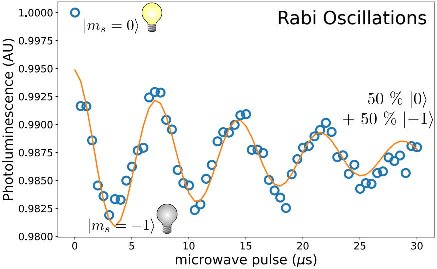


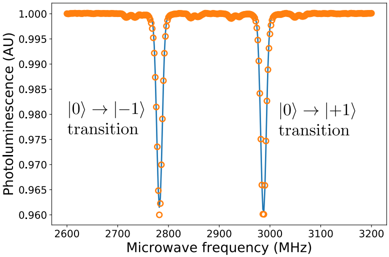
- $|0\rangle$ state brighter than $|\pm 1\rangle$ state by ~ 30 %
- polarization in $|0\rangle$ state of ~ 80 % (equivalent to ~ 65 μK)

- Longitudinal lifetime $T_1 \sim 5$ ms (phonons)
- Dephasing time $T_2^* \sim 1$ μs (magnetic noises)

Spin manipulation



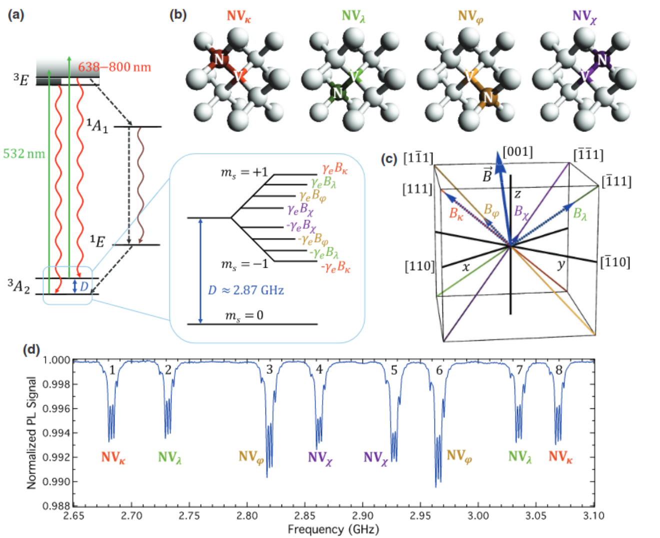




Optically Detected Magnetic Resonance (ODMR)

Summary : Magnetometry with NV centers

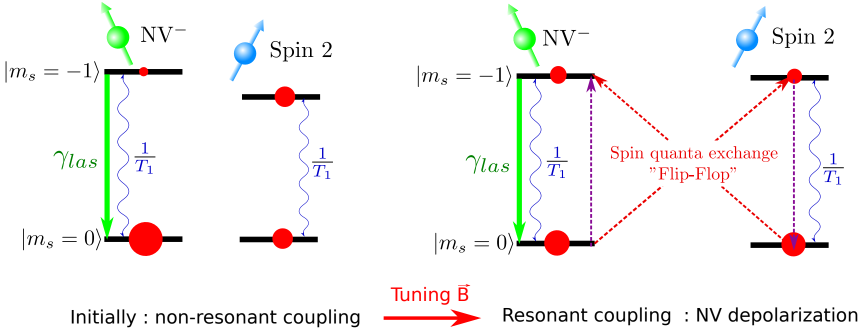
SCHLOSS, BARRY, TURNER, and WALSWORTH PHYS. REV. APPLIED **10**, 034044 (2018)



Principle of spin cross-relaxation (CR)

Dipole-dipole coupling Hamiltonian :

$$\mathcal{H}_{dd} \approx -\frac{\mu_0}{4\pi r^3}\vec{\mu_1}\cdot\vec{\mu_2} \quad \propto \frac{\hat{\vec{S}}_1 \cdot \hat{\vec{S}}_2}{r^3}$$

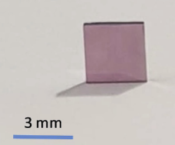


Detection of dark spins in CVD sample

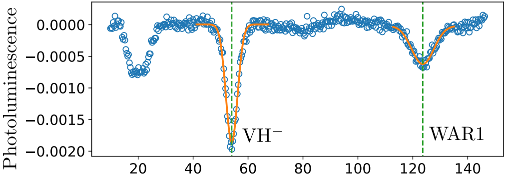
Optical detection of paramagnetic defects in diamond grown by chemical vapor deposition

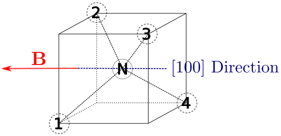
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet

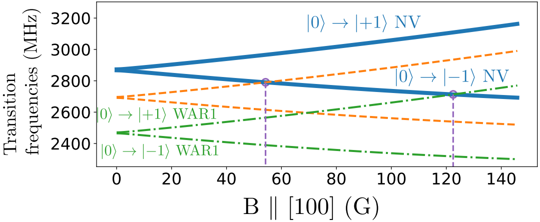
Phys. Rev. B **103**, L100411 – Published 24 March 2021



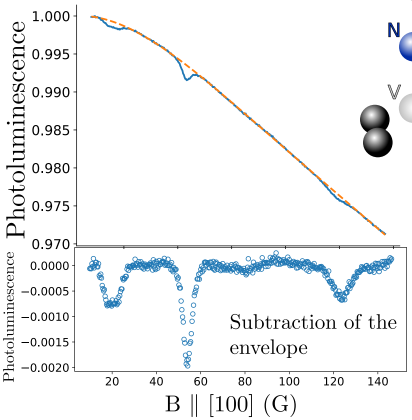
CVD sample from Alexandre Tallaire

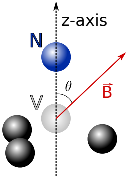






Side-note : effect of transverse field on the PL





Ground level spin Hamiltonian

$$\hat{\mathcal{H}}_s = DS_z^2 + \gamma_e \mathbf{B} \cdot \hat{\mathbf{S}}$$

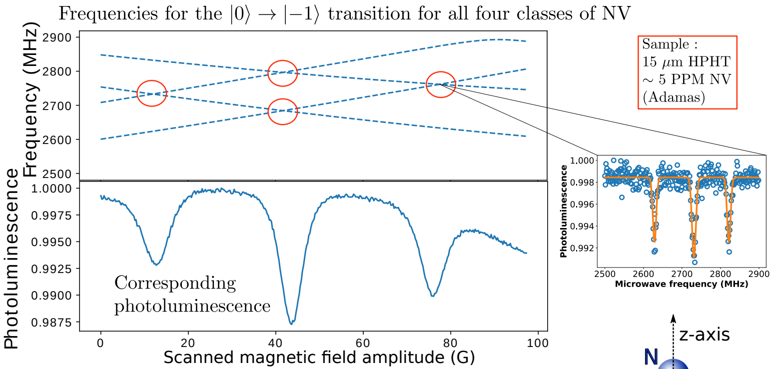
$D = 2.87\,\mathrm{GHz}$ and $\gamma_e = 2.8\,\mathrm{MHz/G}$

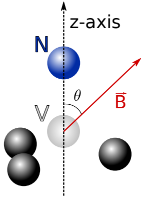
$$\mathcal{H}_s = \begin{pmatrix} D - \gamma_e B\cos\theta & \gamma_e B\sin\theta & 0 \\ \gamma_e B\sin\theta & 0 & \gamma_e B\sin\theta \\ 0 & \gamma_e B\sin\theta & D + \gamma_e B\cos\theta \end{pmatrix}$$

The transverse magnetic field causes state mixing which makes the optical polarization less effective.

→ The photoluminescence decreases with the magnetic field (for non-aligned spins)

NV-NV Cross-Relaxation

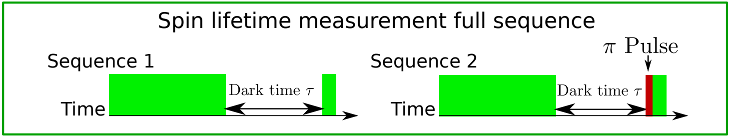


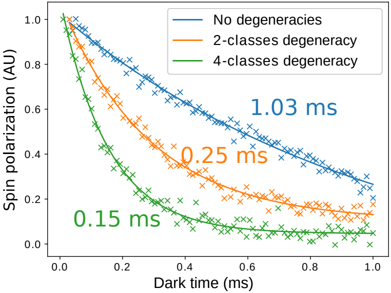


By scanning an exernal magnetic field we can create NV-NV resonances between the four classes.

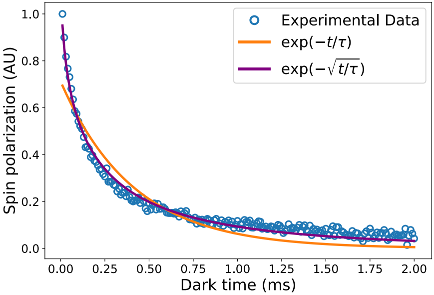
Each NV-NV resonance is met with a sharp drop in the total photoluminescence (for dense NV ensemble)

NV-NV Cross-Relaxation : spin lifetime





Spin lifetime as a function of the number of classes at resonance



Spin lifetime profile (exponential Vs stretch exponential)

Origin of the NV-NV cross-relaxation

Why is it weird ? Because the flip-flop process is spin preserving $\rightarrow$ They should not change the total spin polarization of the NV ensemble.

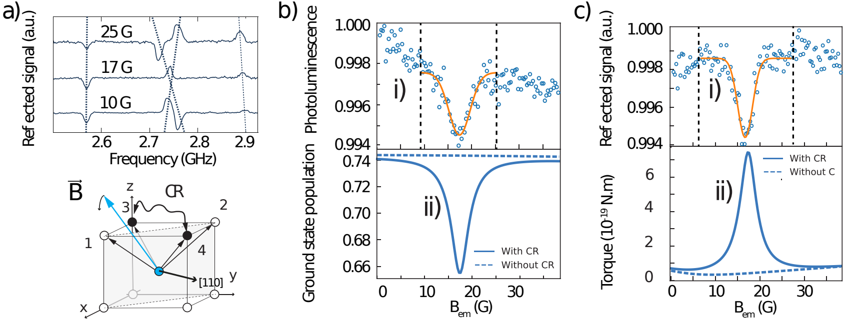
Hypotheses for the origin of the NV-NV Cross-relaxation :

- Spin diffusion to unpolarized spins (out of the laser spot) $\rightarrow$ The numbers are off by several order of magnitudes & it still works with nano-diamond
- Superradiance $\rightarrow$ effect independent of temperature
- Fluctuators : Some NV have their spin heavily depolarized by tunneling in and out of nearby sites
- Polarization of the laser ?

Mechanically detected Cross-Relaxation

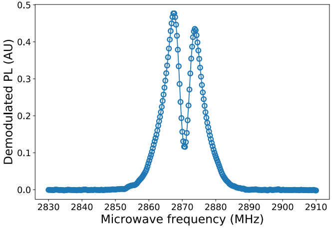
Magnetic torque enhanced by tunable dipolar interactions

C. Pellet-Mary, P. Huillery, M. Perdriat, and G. Hétet
Phys. Rev. B **104**, L100411 – Published 17 September 2021



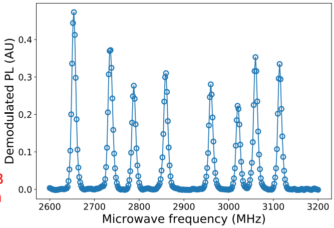
Principle of the magnetometer

ODMR spectrum with microwave modulated in amplitude
(they point upward instead of downward)



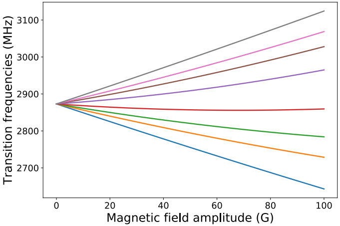
$\vec{B} = 0$

Increasing B in a random direction

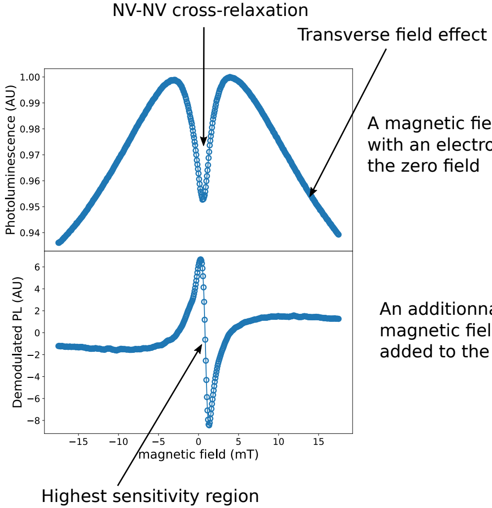


$\vec{B} \approx 100$ G

As the magnetic field increases, the classes get out of resonance from each other
→ They get more polarized
→ The photoluminescence increases



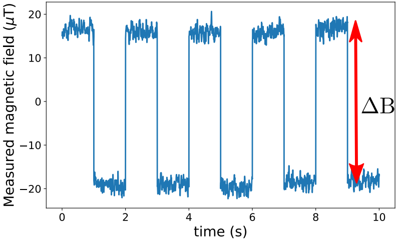
Low field dependence of the PL

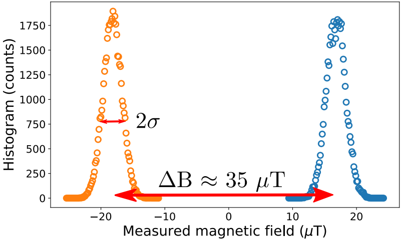


A magnetic field B0 is scanned with an electromagnet around the zero field

An additionnal modulated magnetic field dB can be added to the electromagnet

Sensitivity of the measure





$$\boxed{\begin{aligned}\text{Sensitivity} &= \sigma\sqrt{\tau_{\text{meas}}} \\ &\approx 100\ \text{nT}/\sqrt{\text{Hz}}\end{aligned}}$$

τ_{meas}: Time to perform one measurement (given here by the low pass filter of the lock-in amplifier)

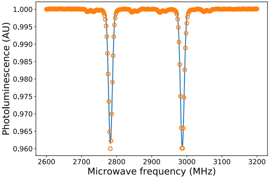
Comparison with other magnetometer

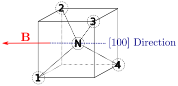
- NV ensemble : ≈ 1 pT$\sqrt{\mathrm{Hz}}$
- Microwave-less NV ensemble : ≈ 1 nT$\sqrt{\mathrm{Hz}}$
- SQUIDs(superconducting circuits)/vapor cells : ≈ 1 fT$\sqrt{\mathrm{Hz}}$

But,

- Based on smaller diamonds (10 μm diamond VS few mm diamonds)
- Does not require well-defined crystal axis Vs magnetic field orientation
  $\rightarrow$ Can use poly-crystalline/powdered samples, diamond flow in solution, etc...
- Better than transverse field by several orders of magnitude

Bonus : Double quantum cross-relaxation in zero-field

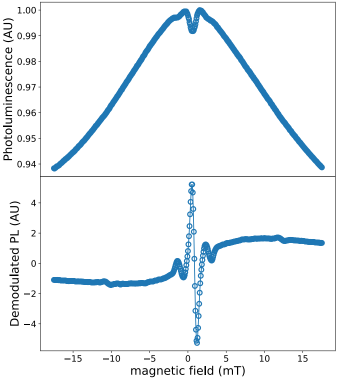


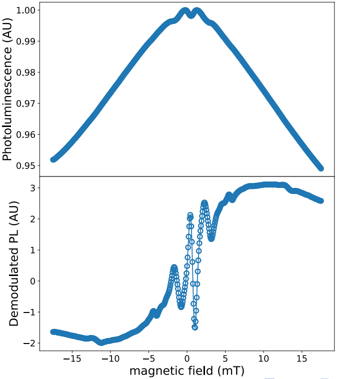


B along 100 : still a dip in zero field ?

→ Double quantum dipolar transition

Ex : $|0\rangle\,|-1\rangle\,\langle+1|\,\langle 0|$ only resonnant on zero field





High variance with the different samples