

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

October 11, 2021

- 1 The NV center
- 2 Probing dark spins with cross-relaxations
- 3 NV-NV Cross-relaxation
- 4 magnetometry with cross-relaxations

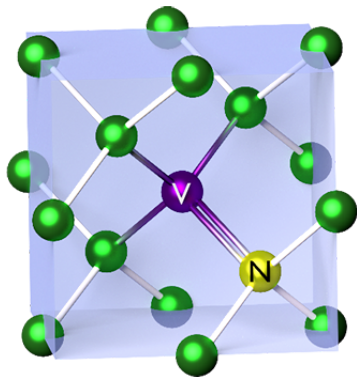
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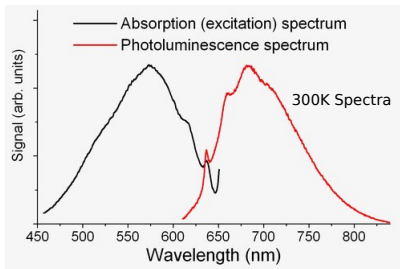
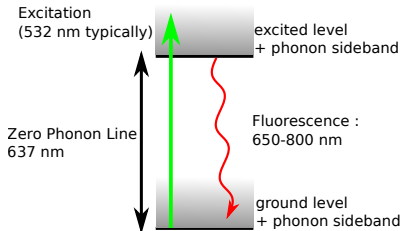
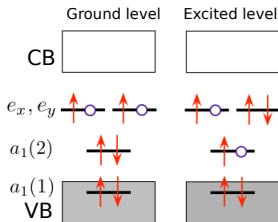
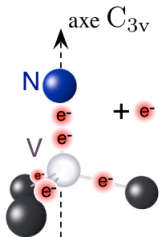
4 magnetometry with cross-relaxations

The Nitrogen Vacancy Center

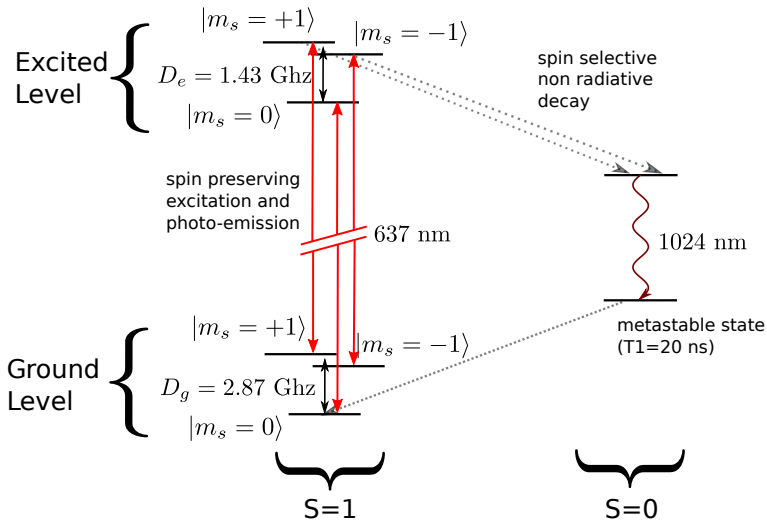


- 0D fluorescent object with ZPL at 638 nm
- Controllable and readable spin at room temperature (!)
- Working with 10^9 emitters (typ.)

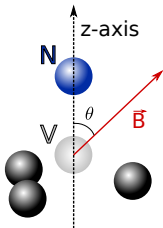
NV center : Optical properties



NV center : 8 levels



NV center : 3 levels

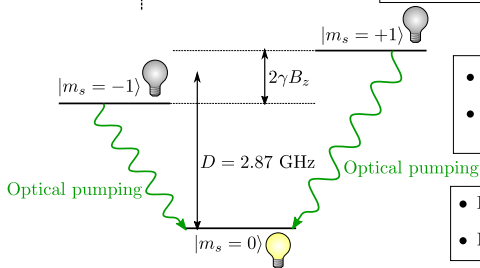


Ground level spin Hamiltonian

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

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

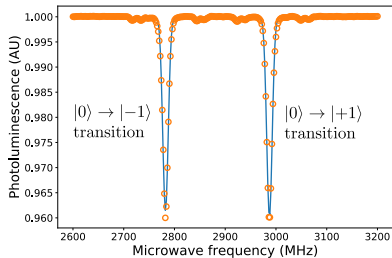
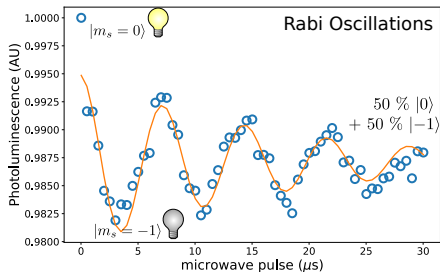
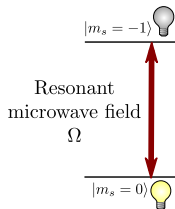
$$\mathcal{H}_s = \begin{pmatrix} D - \gamma_e B \cos \theta & \gamma_e B \sin \theta & 0 \\ \gamma_e B \sin \theta & 0 & \gamma_e B \sin \theta \\ 0 & \gamma_e B \sin \theta & D + \gamma_e B \cos \theta \end{pmatrix}$$



- $|0\rangle$ state brighter than $|\pm 1\rangle$ state by $\sim 30\%$
- polarization in $|0\rangle$ state of $\sim 80\%$ (equivalent to $\sim 65 \mu\text{K}$)

- Longitudinal lifetime $T_1 \sim 5 \text{ ms}$ (phonons)
- Dephasing time $T_2^* \sim 1 \mu\text{s}$ (magnetic noises)

Spin manipulation

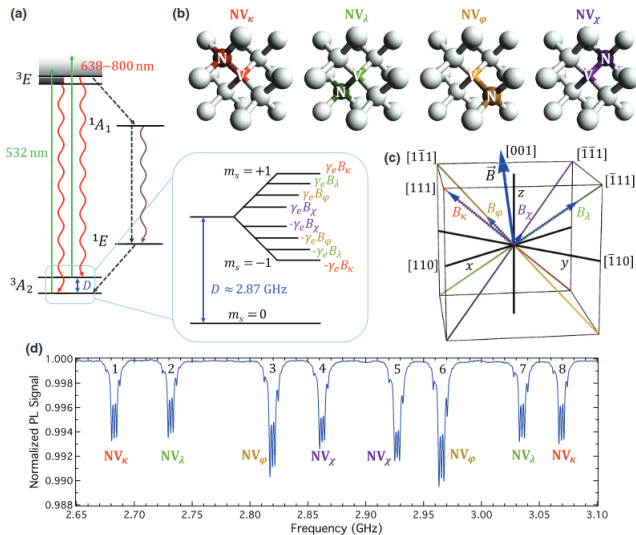


Optically Detected
Magnetic Resonance
(ODMR)

Summary : Magnetometry with NV centers

SCHLOSS, BARRY, TURNER, and WALSWORTH

PHYS. REV. APPLIED **10**, 034044 (2018)



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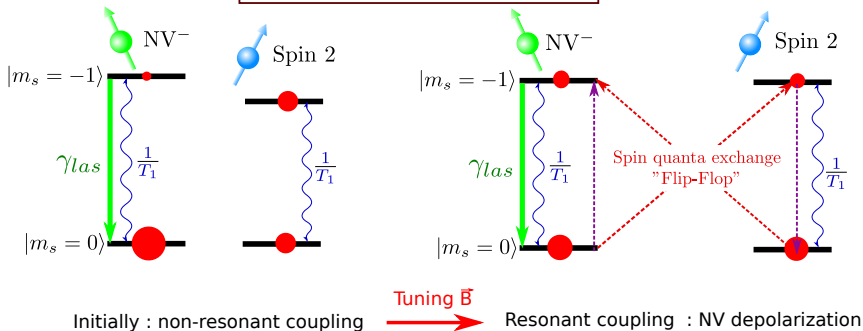
3 NV-NV Cross-relaxation

4 magnetometry with cross-relaxations

Principle of spin cross-relaxation (CR)

Dipole-dipole coupling Hamiltonian :

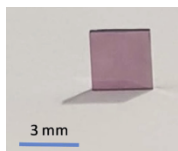
$$\mathcal{H}_{dd} \approx -\frac{\mu_0}{4\pi r^3} \vec{\mu}_1 \cdot \vec{\mu}_2 \propto \frac{\hat{S}_1 \cdot \hat{S}_2}{r^3}$$



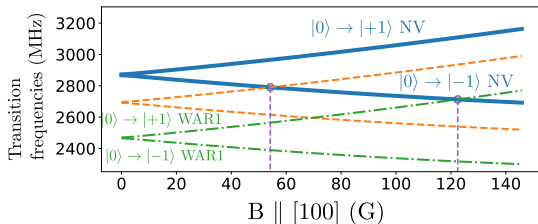
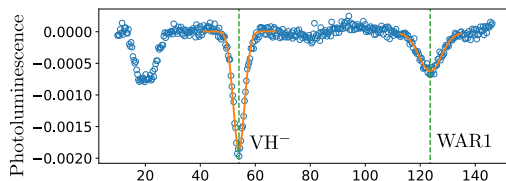
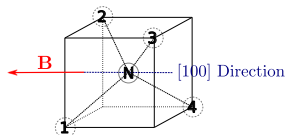
Detection of dark spins in CVD sample

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

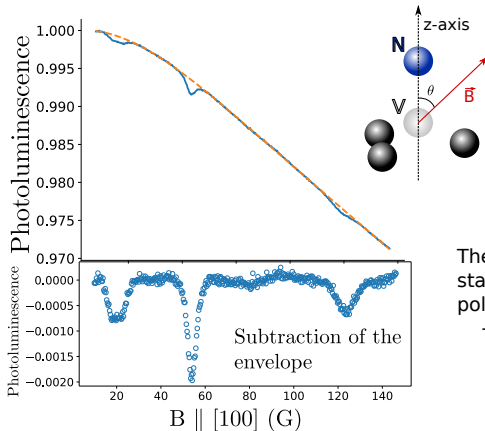
C. Pellet-Mary, P. Huillery, M. Perdriat, A. Tallaire, and G. Hétet
Phys. Rev. B **103**, L100411 – Published 24 March 2021



CVD sample from
Alexandre Tallaire



Side-note : effect of transverse field on the PL



Ground level spin Hamiltonian

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

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

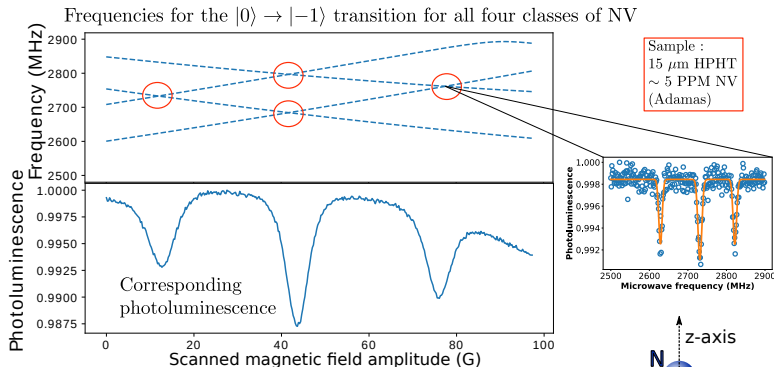
$$\mathcal{H}_s = \begin{pmatrix} D - \gamma_e B \cos \theta & \gamma_e B \sin \theta & 0 \\ \gamma_e B \sin \theta & 0 & \gamma_e B \sin \theta \\ 0 & \gamma_e B \sin \theta & D + \gamma_e B \cos \theta \end{pmatrix}$$

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)

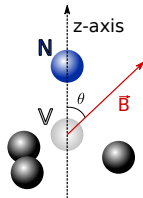
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NV-NV Cross-Relaxation

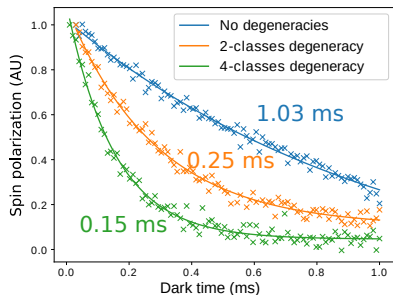
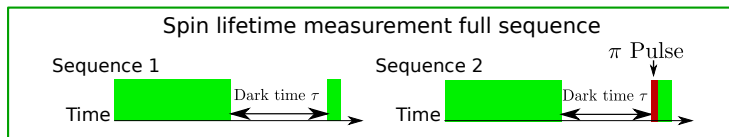


By scanning an external magnetic field we can create NV-NV resonances between the four classes.

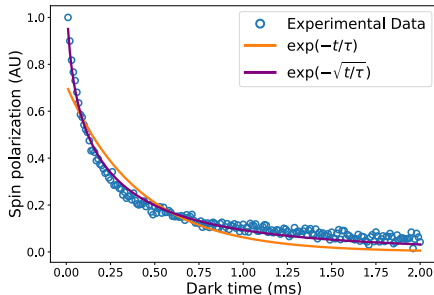
Each NV-NV resonance is met with a sharp drop in the total photoluminescence (for dense NV ensemble)



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
→ They should not change the total spin polarization of the NV ensemble.

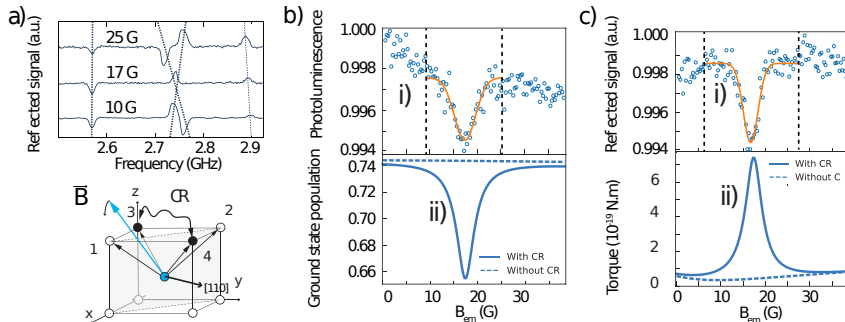
Hypotheses for the origin of the NV-NV Cross-relaxation :

- Spin diffusion to unpolarized spins (out of the laser spot)
→ The numbers are off by several order of magnitudes & it still works with nano-diamond
- Superradiance → 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

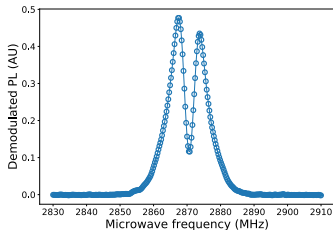
C. Pellet-Mary, P. Huillery, M. Perdriat, and G. Hétet
Phys. Rev. B **104**, L100411 – Published 17 September 2021



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Principle of the magnetometer

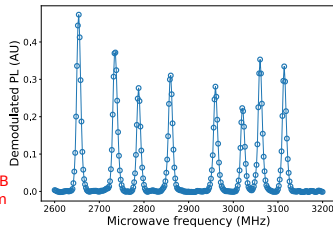
ODMR spectrum with microwave modulated in amplitude
(they point upward instead of downward)



$\vec{B} = 0$

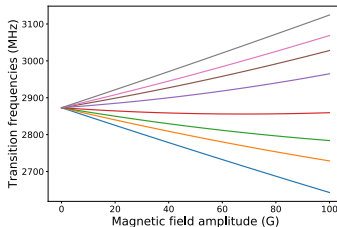


Increasing B
in a random
direction

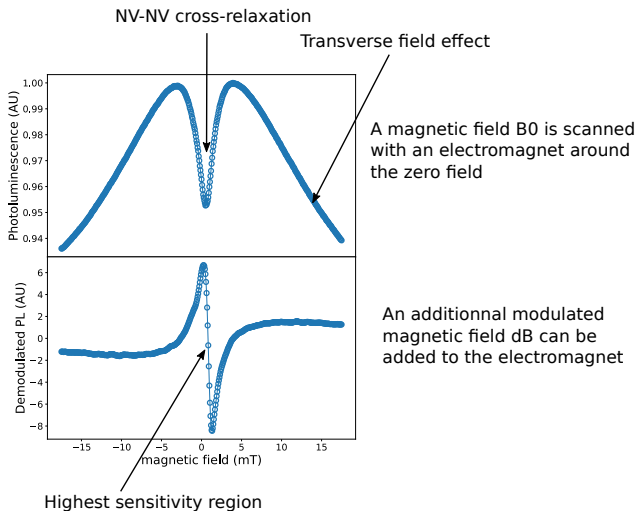


$\vec{B} \approx 100 \text{ G}$

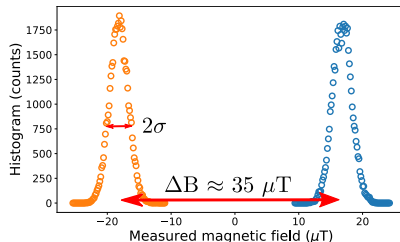
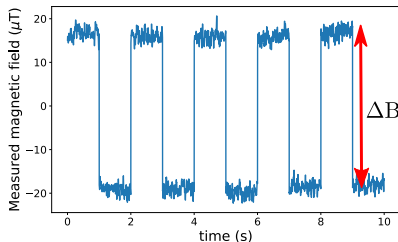
As the magnetic field increases, the classes get out of resonance from each other
→ They get more polarized
→ The photoluminescence increases



Low field dependence of the PL



Sensitivity of the measure



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

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

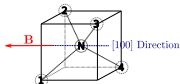
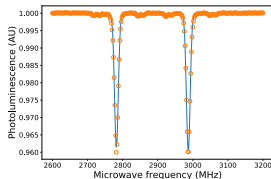
Comparison with other magnetometer

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

But,

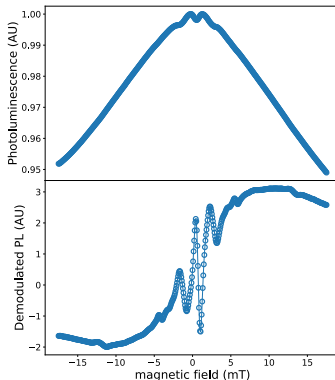
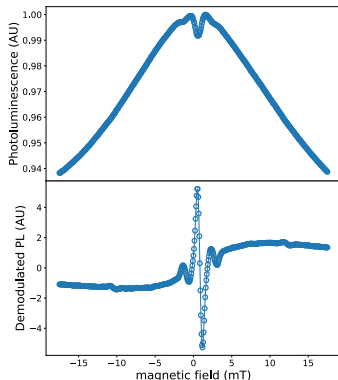
- Based on smaller diamonds (10 μm diamond VS few mm diamonds)
- Does not require well-defined crystal axis Vs magnetic field orientation
→ Can use poly-crystalline/powdered samples, diamond flow in solution, etc...
- Better than transverse field by several orders of magnitude

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 resonant on zero field



High variance with the different samples