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C'est trop bien

Blabla sur les NV, les ensembles etc.

In this letter/article, we characterize the depolarization of the spins observed for dense ensemble of NV centers in zero magnetic field and its potential application for DC magnetometry. While the main mechanism behind the depolarization, the lift in the degeneracy between the four classes of NV centers, is already well studied and can be exploited in a microwave-less vector magnetometry protocol, we found two other depolarization mechanisms specific to the zero-field region which could play an important role in the low-field magnetometry protocol.

The T_1 depolarization dynamics of single or sparse NV centers at room temperature is dominated by a two-phonon Raman process, which depends on the crystal lattice temperature but does not (?) depend on the external magnetic field. However, it has been observed that dense NV centers ensemble have an additional spin lifetime contribution which depends greatly on the magnetic field and not on the temperature. This effect has been attributed to cross-relaxation between the NV centers through dipole-dipole coupling. Some inhomogeneity between the NV centers is further needed in order to explain the depolarization of the spin ensemble.

The main signature of the spin depolarization in low field is the characteristic dip in photoluminescence (PL) observed only for high density ($\gtrsim 1$ ppm) of NV centers, as shown on fig 1. The decrease in the spins' lifetime in zero field makes the optical polarization scheme of the NV centers less effective and therefore reduce the population of the bright $|0\rangle$ spin state. The reason for the decrease in PL at higher magnetic field values is the mixing of the bright spin state $|0\rangle$ to the darker state $|-1\rangle$ induced by the transverse magnetic field. This effect does not modify (on first approximation) the spins' lifetime, and is common to both dense and sparse samples.

The spin lifetime protocol used here is described in Fig. 2 and is based on previous similar experiments. It consists in a classical pump-probe measurement where the spins are first polarized in the $|0\rangle$ state by a green laser, and read-out optically after a variable dark time τ. Using only this sequence can result in artifacts in the final T_1 signal due to charge state transfer in the dark, particularly for dense ensemble. It is therefore convenient to repeat the sequence with an additional π pulse right before the spin read-out to project the remaining $|0\rangle$ polarization into a darker $|+1\rangle$ or $|-1\rangle$ state. By subtracting the result of the two sequences, we select only the

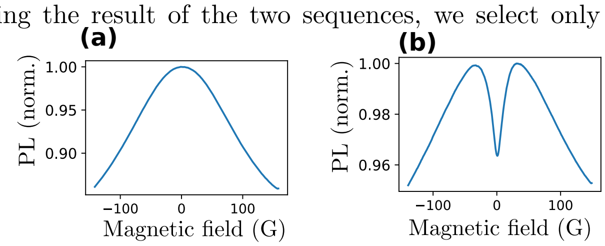

FIG. 1. Photoluminescence from NV centers as a function of an external magnetic field randomly oriented for two samples : (a) a CVD sample containing ≈ 4 ppb NV^- centers, (b) an HPHT sample containing ≈ 3 ppm NV^- centers

spin-dependent part of the signal, with the added benefit of being able to select a specific class of NV centers.

The ensemble spin lifetime is then fitted with a formula which include both an exponential lifetime T_1^{exp} and a stretch exponential lifetime T_1^s :

$$S(\tau) = A\exp\left(-\frac{\tau}{T_1^{\mathrm{exp}}} - \sqrt{\frac{\tau}{T_1^s}}\right), \tag{1}$$

We assume that the exponential part of the lifetime is defined by the phonons, and the stretch part by the coupling between NVs, as detailed below. In order to get a comparable ... between each measurement, we decided to fix the value of the exponential lifetime in the fitting process, leaving only the stretch lifetime as a free parameter. For Fig.2, 3, 4 and 6, the chosen exponential lifetime was $T_1^{\mathrm{exp}} = 3.62ms$.

The additional spin depolarization observed

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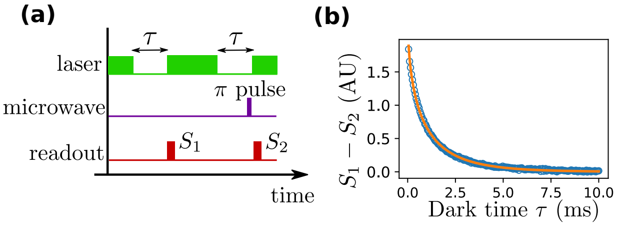

FIG. 2. Spin T_1 measurement protocol. (a) Sequence used to measure the spin lifetime : the laser is turned off for a time τ twice and the spin state is read at the end of the dark time by measuring the initial photoluminescence when the laser is turned back on. A microwave resonant with one of the spin transition is used in one of the sequence to project the $|0\rangle$ spin sate in either $|+1\rangle$ or $|-1\rangle$. The sequence result is the subtraction $S_1 - S_2$. (b) Measurement of an ensemble spin lifetime, fitted with an expression $A\exp\left(-\frac{\tau}{T_1^{\exp}} - \sqrt{\frac{\tau}{T_1^s}}\right)$

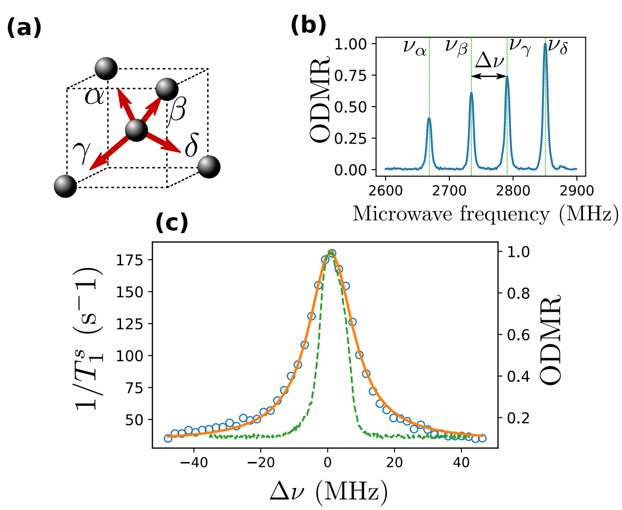

FIG. 3. Tuning resonant coupling through magnetic field orientation. (a) Sketch of the four possible orientations ("classes") in a single crystal diamond lattice. (b) ODMR spectrum showing four $|0\rangle \rightarrow |-1\rangle$ resonances corresponding to the four spin classes. The detuning $\Delta\nu$ between the classes β and γ was controlled by changing the orientation of the external magnetic field.(c) Stretch part of the lifetime decay for the spins resonant with ν_γ as a function of the detuning $\Delta\nu$ (blue circles), fitted by a Lorentzian with half width at half maximum 8.04 MHz . Green dashed line correspond to the ODMR width of a single class stretched by a factor $\sqrt{2}$, approximating the overlap of the two classes (see SI).

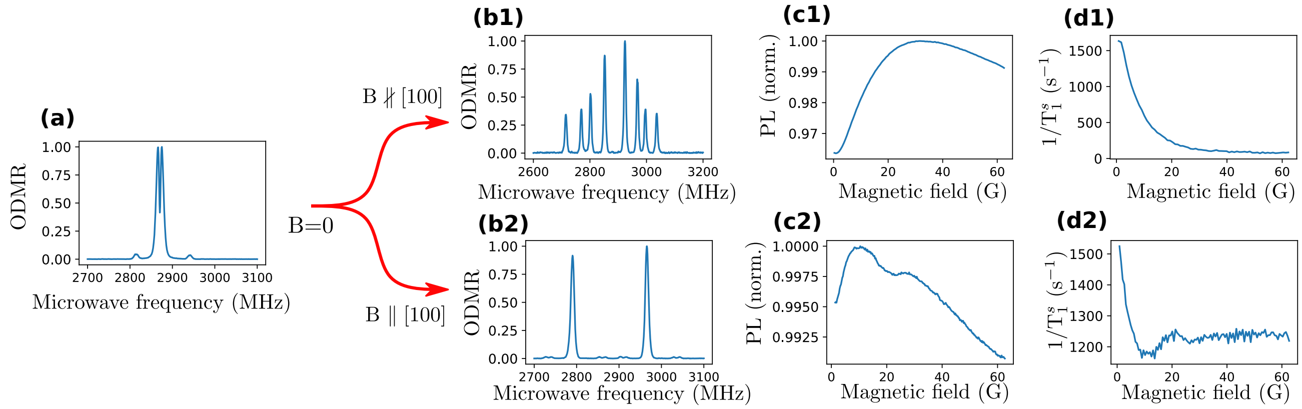

FIG. 4. Dependency of the magnetic field angle for the zero field depolarization. (a) ODMR spectrum in zero field. (b) ODMR spectrum for a magnetic field ≈ 60 G. The field is aligned (b2) or misaligned (b1, ≈ 24° of misalignment) with the crystalline [100] direction. (c) Normalized photoluminescence of the NV^- ensemble as a function of the magnetic field amplitude for the same field orientations as fig (b). (d) Stretch part of the lifetime decay as a function of the magnetic field amplitude for the same field orientations as fig (b).

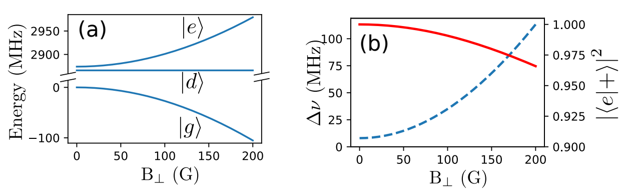

FIG. 5. Simulated eigenstates of the spin Hamiltonian in the presence of purely transverse magnetic field. (a) Energies of the three eigenstates $|g\rangle$, $|d\rangle$ and $|e\rangle$ as a function of the magnetic field amplitude. (b) Blue dashed curve : frequency detuning between the two transitions $|g\rangle \leftrightarrow |d\rangle$ and $|g\rangle \leftrightarrow |e\rangle$. Plain red curve : projection of $|e\rangle$ on $|+\rangle = (|{+1}\rangle + |{-1}\rangle)/\sqrt{2}$ as a function the magnetic field. Is equal to the projection of $|g\rangle$ on $|0\rangle$.

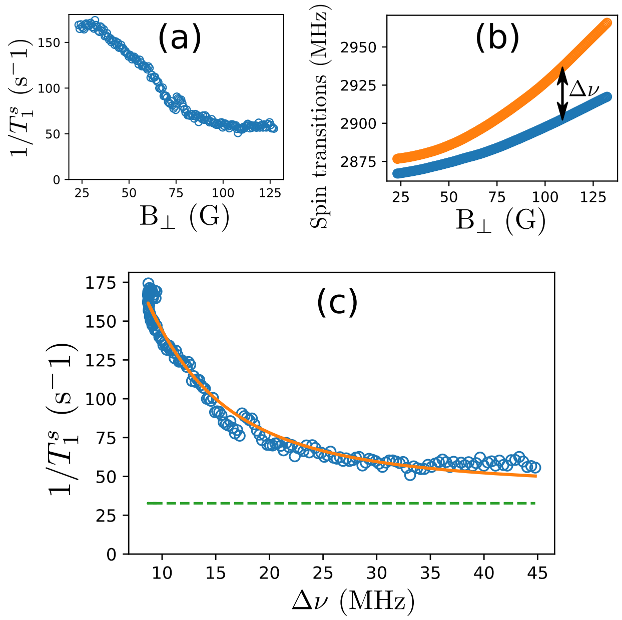

FIG. 6. Modification of the stretch lifetime in the presence of purely transverse magnetic field. (a) Stretch component of the ensemble lifetime as a function of the field amplitude. (b) Measured transition frequencies through ODMR spectrum. (c) Stretch component of the lifetime as a function of the frequency detuning between the two transistions (blue circles), fitted by a Lorentzian centered in $\Delta\nu = 0$ with half width at half maximum 8MHz. The green dashed line correspond to the lifetime of a single class aligned with the magnetic field.