

Optical detection of hydrogen paramagnetic defects in diamonds

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We observe cross-relaxation amongst closely packed negatively charged nitrogen-vacancy centers (NV^-) that involve dipolar interaction between NV^- centers and paramagnetic defects. Specifically, we observe dipolar coupling between pairs of NV^- centers and between single NV^- centers and $NV^- -^{13}C$ pairs as well as novel optical cross-relaxation with hydrogen-vacancy complexes. These processes are detected using magnetic fields scans along the $\langle 100 \rangle$ crystalline direction of diamond. Contrary the more commonly chosen $\langle 111 \rangle$ direction, $\langle 100 \rangle$ scans select dipolar couplings involving the exchange of two quanta of spin-projection. The technique can be used to detect the degree of polarisation of ^{13}C atoms at room temperature.

The precise control of the spin of negatively charged nitrogen-vacancy (NV^-) centers in diamond has given rise to a wealth of applications in nanoscale sensing, quantum information science etc.. One major reason for the interest in NV^- centers is that they can be optically polarized at room temperature. This property can be employed to manipulate and to detect other dark spins inside or outside the diamond.

Cross relaxation. Most of the measurements are done at the ESLAC/GLSAC. OORT, BUDKER, HOLLENBERG, BAJAJ, MANSON ?

Polarisation of ^{13}C at low fields : PINES, MERILES. Theory GALI. Detection at low fields was also demonstrated [1? -7]. It was suggested that spin relaxation beyond the phonon-relaxation timescales comes from electron tunneling between nearby impurities. Dipolar coupling amongst closely packed NV^- was used further to demonstrate many body effects such as critical thermalisation, or Dicke time crystals or beyond Heisenberg limited sensing (Lukin).

To the best of our knowledge, the process was however not employed to detect other impurities.

D. SUTER. However, ^{13}C was not detected. The main reason is the predominance of single-quantum processes. ^{13}C detected using B field scans ? nop. Here, we characterize the cross-relaxation of high-density NV^- ensembles with a focus on double quantum resonances.

Fig. 4 shows the NV^- Photoluminescence as a function of magnetic field direction, referenced by azimuthal and polar angles (ϕ, θ) . The PL is seen to drop for particular values of the angle. The symmetric pattern suggests that the diamond crystalline main directions play a role. Because of the anisotropy of the NV^- center, see 1, the NV^- PL drop can indeed be a signature of the cristal direction. The drop of the PL along the cristal axis was already observed by many groups. LUKIN, HEMMER, BUDKER. JARMOLA. OORT [1? -7]

Charge transfer processes :

IONISATION 2 photon Photoionisation from NV^- to NV^0 . OR Tunneling (Manson/Degen) : NV^- excited state ionizes via tunneling of electrons to Ns^+ .

RECHARGING : $NV^0 + N_0 \rightarrow NV^- + N^+$ (in the dark). It can occur via tunneling of electrons among closely

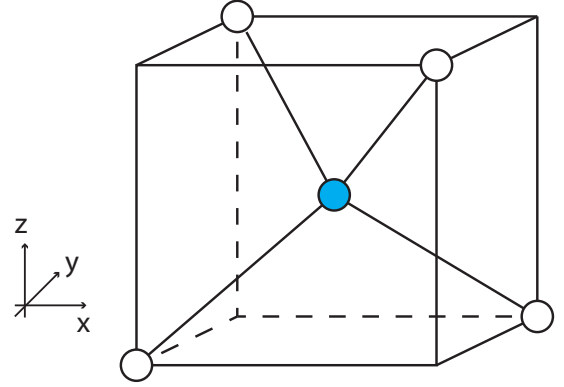


FIG. 1. a) Schematics showing the tetrahedron in one of the interstitial sites of the diamond structure. The NV^- centers can only be in one of the four depicted orientations.

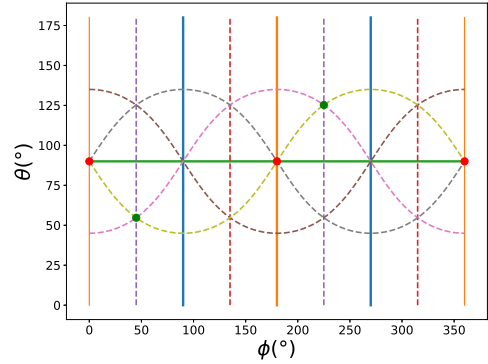


FIG. 2. a) Graph (theory) showing the locus of the degeneracies of the 4 NV^- centers electronic spins resonances in the (θ, ϕ) parameter space. See text for details.

packed NV^- s.

Each of the lines is very well approximated by a Gaussian curve.

Fig. 5 shows measurements of the T_1 / Tunneling is strong in the presence of nitrogen impurities. To remove this effect, we apply a microwave....

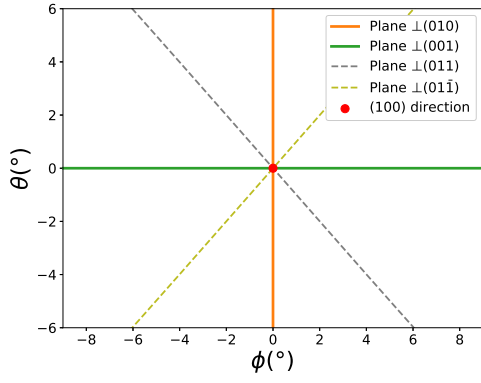


FIG. 3. a) Zoom on the $\langle 100 \rangle$ direction.

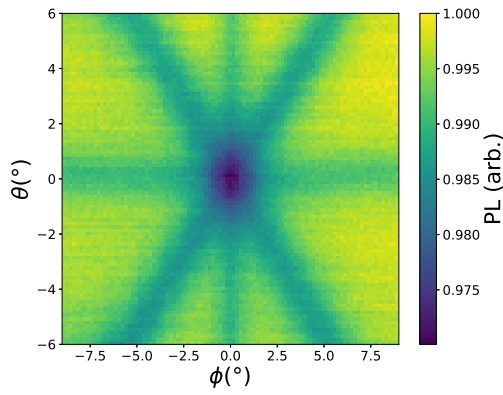


FIG. 4. a) NV Photoluminescence as a function of magnetic field direction, referenced by azimuthal and polar angles (ϕ, θ) .

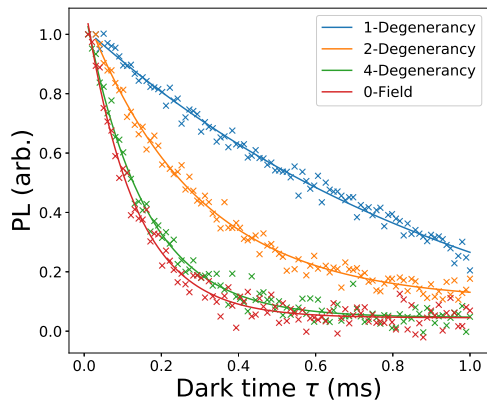


FIG. 5. a) Lifetime saturation theory

Two peaks cannot be explained.

Hypotheses that did not work.

NV^0 , $P1$, $13C$ other shells, NVN (H3) center (diamagnetic in ground state), paramagnetic only in excited state

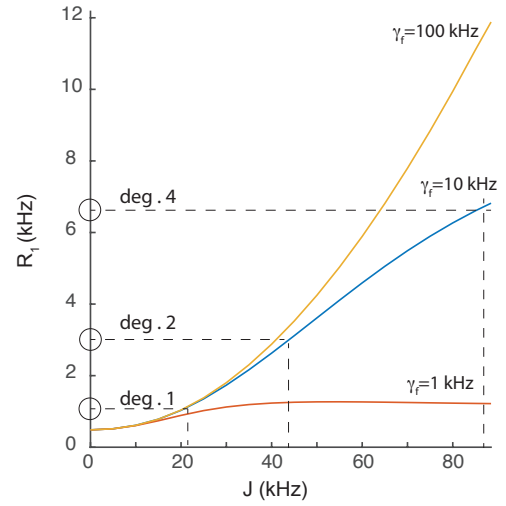


FIG. 6. a) Lifetime saturation theory

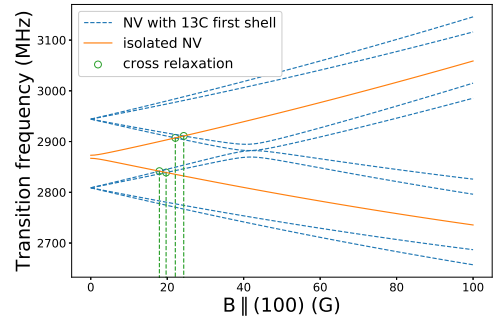


FIG. 7. Transitions of NV- ^{13}C and NV.

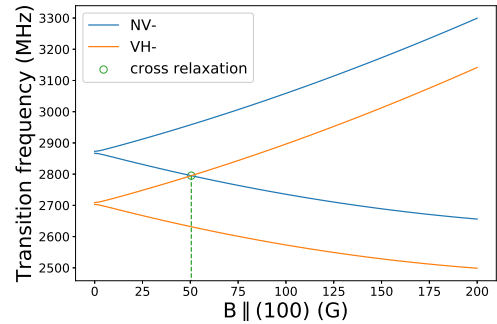


FIG. 8. Transitions of VH- and NV.

but requires UV light. No other species were observed in the optical spectrum or EPR.

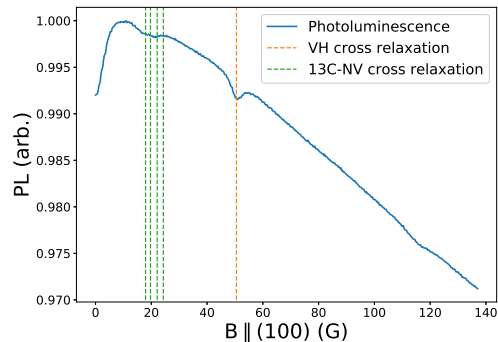


FIG. 9. NV Photoluminescence as a function of a magnetic field scanned along the 100 direction.

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