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Optical polarization of molecules using nitrogen vacancy centres in diamond Benedikt Tratzmiller Institut für Theoretische Physik und IQST, Albert-Einstein-Allee 11, Universität Ulm, 89081 Ulm, Germany

Abstract

This Poster summarises my Master thesis [1] that I submitted in December 2016 at Ulm University. Here I focus on the most important part of my thesis, the development of an error-robust, fast and efficient polarisation transfer protocol that is beneficial for many applications termed PulsePol. This technique is particularly useful for dynamic nuclear polarisation (DNP) with colour centres in diamond, where it has the potential to increase the signal in magnetic resonance imaging (MRI) by a factor > 10000 without the disadvantages of other methods. Furthermore PulsePol is useful for the error-robust initialisation of quantum registers and nuclear spin simulators.

This poster covers motivation, description and development of the PulsePol sequence as well as the theoretically simulated and experimentally confirmed advantages.

Background: MRI and DNP

Magnetic resonance imaging (MRI): Medical images without nuclear radiation, but **low signal to noise ratio** and the resulting low resolution. Origin: low nuclear magnetisation achievable at room temperature, related to a Boltzmann distribution (room temperature, magnetic field B = 3T, hydrogen spins) \rightarrow Only one of 100000 spins is responsible for the contrast.





Solution: Polarise the nuclear spin bath significantly beyond thermal polarisation (**hyperpolarisation**), e.g. **dynamic nuclear polarisation (DNP)** (transferring polarisation from an electron spin to nuclear spins) Established: thermally polarised electron spins at cryogenic temperatures and high magnetic fields [2]. More flexible: colour centres in diamond

New principle: Pulsed polarisation

Idea:

- Resonance condition does not depend on Rabi frequency (now: evolution time, easier to control)
- Increase robustness to systematic experimental errors (detuning, Rabi frequency error, phase error) with a pulse sequence (similar to sensing sequences like XY-4 etc.)

Criteria for a robust polarisation sequence:

- 1. Correct detuning and Rabi errors (free evolutions and pulses)
- 2. Basis changes between $S_{x/y}$
- 3. Filter functions shifted relative to each other, ideally by a phase $\pi/2$



Nitrogen Vacancy (NV) centres



The Nitrogen Vacancy (NV) centre is a crystal defect in diamond that consists of a nitrogen atom and an adjacent lattice vacancy.
Its spin 1 ground state has many favorable properties for DNP
Easy to initialise and read out

• Stable, good T_1 and T_2 times even at room temperature

Pulsed polarisation sequences: PulsePol

Based on those criteria, I analytically derived the PulsePol sequence consisting of π and $\pi/2$ pulses around the X- and Y-axis and free evolution periods (0 driving amplitude) for a time $\tau/4$. **Resonance condition** (ω_L : nuclear Larmor frequency, *n*: odd integer. Fastest transfer for n = 3):



Controllable with microwave pulses
Dipole-dipole coupling to nuclear spins

Due to the large zero field splitting $D = (2\pi) 2.87$ GHz, the effective energy splitting between the NV eigenstates depends critically on the NV orientation. Therefore DNP with NV centres in nanodiamond ensembles is an unmet challenge.



Problems of existing DNP methods

Challenge for DNP: overcome different energy splittings for electron and nuclear spin (factor ${\sim}2000$).

Existing methods for DNP are either slow or prone to experimental errors (Highest practical impact: frequency mismatch (detuning) errors) \rightarrow reduced DNP efficiency

NV centres: orientation dependent energy splitting.

NOVEL (Nuclear spin orientation via electron spin locking) [3]. Effective Hamiltonian (S(I): electron (nuclear) spin operators):

$$H_{\text{NOVEL}} = \frac{A_{\perp}}{2} \left(S_x I_x + S_y I_y \right) = \frac{A_{\perp}}{4} \left(S_+ I_- + S_- I_+ \right)$$



Fast polarisation transfer, but not very robust against errors.

Spin locking, $X: \Omega = \omega_L$

 $\wedge \wedge \wedge$

 $\frac{\pi}{2}$

Transfer simulation: Detuning $\Delta = (2\pi)0.5$ MHz (blue

Effective Hamiltonian (S(I): electron (nuclear) spin operators):

$$H_{\mathrm{PP}} = rac{2(2+\sqrt{2})A_{\perp}}{3\pi} \left(S_x I_x + S_y I_y
ight) pprox 0.72 H_{\mathrm{NOVEL}}$$





Fast, robust and efficient polarisation transfer with PulsePol.

Transfer simulation: Detuning errors $\Delta = 0.1\Omega_0 = (2\pi)5MHz$ (dashed blue line) and Rabi frequency errors $\delta\Omega = 0.1\Omega_0 = (2\pi)5MHz$ (dash-dot green line) have a very small effect.



Left: Robustness of the polarisation transfer of one electron spin excitation to five nuclear spins for the PulsePol (insert: NOVEL) scheme for $\omega_L = 2$ MHz, $\Omega_0 = (2\pi)50$ MHz and a spin bath with an average $\langle A_{\perp} \rangle = 30$ kHz.

Right: Experimentally measured detuning resistance of PulsePol (red) and NOVEL (blue). Solid lines are the result of a simulation of a comparable nuclear spin bath with no free parameters. [4]

dashed) or Rabi frequency error of 2% (green dashdotted) \rightarrow transfer efficiency <10%. Further Improvement: Composite Pulses, shaped pulses etc.

Conclusion

- New principle: PulsePol to polarise nuclear spins
- Mathematically and physically correct, experimentally verified
- Various quantum applications: MRI-DNP, initialisation of quantum registers and nuclear spin simulators



[1] Tratzmiller, B. "Optical polarization of molecules using nitrogen vacancy centres in diamond". Master Thesis at Ulm University (2016).

[2] Ardenkjær-Larsen, Jan H., et al. "Increase in signal-to-noise ratio of> 10,000 times in liquid-state NMR." Proceedings of the National Academy of Sciences 100.18 (2003): 10158-10163.

[3] Henstra, A., et al. "Nuclear spin orientation via electron spin locking (NOVEL)." Journal of Magnetic Resonance (1969) 77.2 (1988): 389-393.

[4] Schwartz, I., Scheuer, J., Tratzmiller, B. et al. "Pulsed polarisation enabled by Hamiltonian engineering", submitted.

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