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PHOTONIC AND NANOMETRIC HIGH-SENSITIVITY BIO-SENSING

DELIVERABLE 1.2 [HUJI, M42]
REPORT ON THE DEMONSTRATION OF OPTIMIZED ADRT COOLING

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OPTIMIZED ADRT COOLING WITH NVS

Work Package 1

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Contents

1 Executive Summary

As deliverable D1.2 we report on our results of using a nitrogen vacancy (NV) center to cool a surrounding nuclear spin bath (comprising of 13C impurities):

- We expand previous work and relate to our theoretical results [1], and study Hartman-Hahn (HH) resonant transfer along with optimized alternating resonant and dissipative coupling (optical pumping).
- **We measure an enhancement of the NV coherence time (its T_2^* time) as an indicator of the bath cooling (polarization).**
- A few open questions remain and are the focus of continued research.

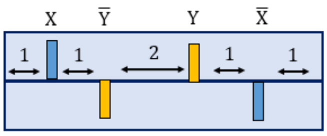
2 Cooling techniques

We have developed several cooling techniques relevant for the hyperpolarization of a spin bath through coupling to NV centers, which are cooled through their intrinsic optical pumping process. These techniques include: (i) the standard approach based on the HH scheme [2], which essentially relies on spin-lock driving of the NV along with the target spins; (ii) a modified approach specifically relevant for nuclear spins at low magnetic fields, extending the standard scheme NOVEL (nuclear orientation via electron spin-locking) by integrating refocusing pulses, which we denote rNOVEL (refocused NOVEL) [3]; (iii) an advanced scheme incorporating polarization transfer with a dense, interacting NV spin ensemble [4]; (iv) an alternating scheme based on resonant transfer and optical pumping cycles as a function of magnetic field, tuning the efficiency of the polarization transfer process near the level anti-crossing.

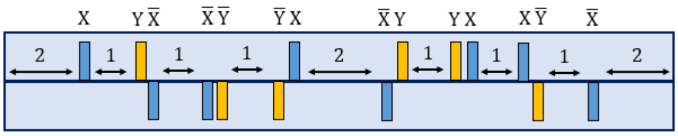
We detail below new approaches developed recently, namely polarization transfer from an interacting spin ensemble and cooling near the level anti-crossing.

2.1 Polarization transfer from an interacting spin ensemble

We studied the quantum many-body dynamics of an interacting open spin system (ensemble of NV centers), coupled to a spin bath (nuclear spins of a target sample). This scenario addresses the basic aspects of PATHOS in terms of system-bath coupling and control, and connects to real-world applications, such as enhanced sensitivity in sensing magnetic samples and hyperpolarization of samples for NMR/MRI.
We analyzed these dynamics theoretically and developed a general framework for the control of interacting spin systems in the average Hamiltonian picture [K. Ben'Attar et. al., PRR 2, 013061 (2020)]. We then utilized this framework to develop and optimize a polarization transfer scheme which combines resonant transfer approaches (such as Hartmann-Hahn) with decoupling protocols. We showed that our approach can improve over existing methods in terms of achievable polarization level and speed of polarization transfer [K. Ben'Attar et. al., in preparation].



(a) WaHuHa



(b) Proposed sequence

Figure 1: Pulse sequences for polarization transfer. (a) Standard WAHUHA sequence. (b) Newly developed sequence optimized for interacting spins, derived through linear programming.

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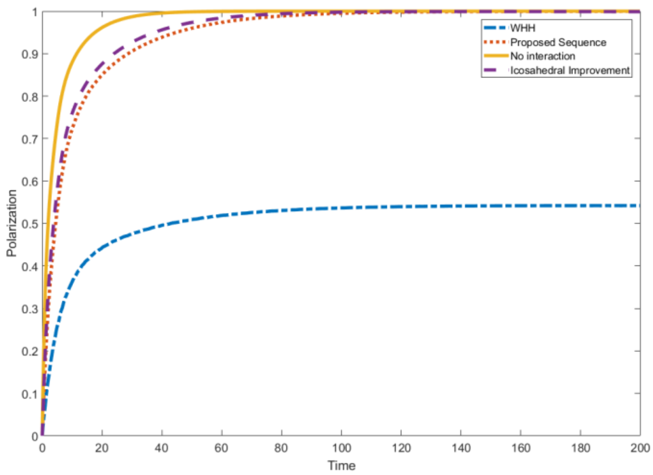

Figure 2: Achieved polarization as a function of time, comparing between the different sequences (non-interacting system, WAHUHA, optimized cubic sequence and the optimized icosahedral sequences). The optimized sequences display a significantly improved efficiency compared to WAHUHA, close to the optimal non-interacting case.

2.2 Polarization transfer near the level anti-crossing

While working on the experimental realization of the NOVEL and rNOVEL schemes (see preliminary results below), we identified a dependence of the inhomogeneous dephasing time T_2^* on the externally applied magnetic field. This dependence essentially presented a difference between the $|0\rangle \leftrightarrow |1\rangle$ and the $|0\rangle \leftrightarrow |-1\rangle$ dephasing times, exhibiting ~50% longer coherence times for the -1 transition compared to the +1 transition for magnetic fields near the excited state level anti-crossing (ESLAC). Therefore we attribute this behavior to the efficient polarization transfer process occurring at the ESLAC due to the resonant condition between the -1 transition of the NV electronic spin and the transition of nearby nuclear spin in the bath.

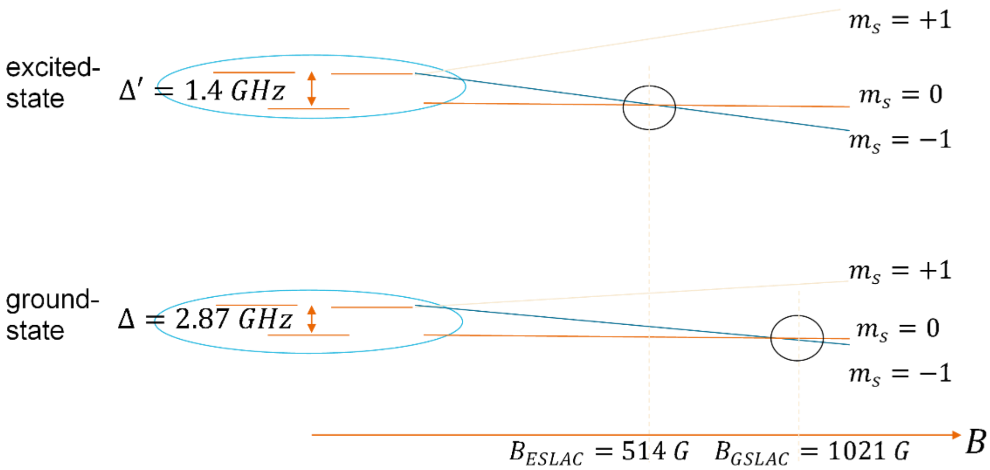

Figure 3: Energy level diagram of the NV electronic spin as a function of magnetic field, indicating the ESLAC and GSLAC.

We expect another resonant transfer to occur near the ground-state level anti-crossing (GSLAC), albeit with somewhat lower efficiency due to the smaller hyperfine coupling. We studied these coherence properties on various NV centers and on different samples, notably a sample purified to have a negligible concentration of nuclear spins (13C impurities). Our results are consistent with the cooling process described above, although a change in the coherence time for the purified sample near the GSLAC is still not understood.
These results are currently being complemented by additional measurements and theoretical analysis and are being prepared for publication.

3 Experimental results

Based on our theoretical analysis and results described above and detailed in the references [1,3,4], as well as previous experimental results [2], we have continued with experimental realizations of the different cooling schemes and regimes using the platform of NV centers in diamond. We describe below our current results on NOVEL and rNOVEL, as well as on cooling near the level anti-crossing.

3.1 NOVEL and rNOVEL

We carried out experiments on polarization transfer based on the NOVEL and rNOVEL schemes, working at different magnetic fields with an emphasis on low fields for which the rNOVEL technique was developed and is optimized.

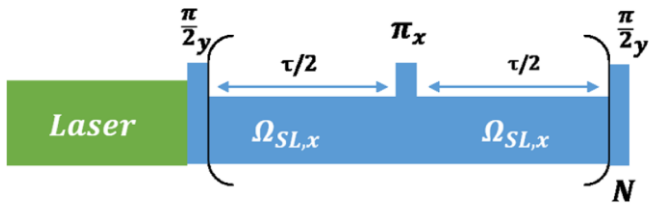

Figure 4: Schematic representation of the rNOVEL sequence. The NOVEL sequence can be derived from it by removing the refocusing π pulse.

While these results indicate the enhancement achieved through this new approach, they were realized on an ensemble of NVs, and therefore the effect is inhomogeneously broadened and less pronounced. We are therefore currently extending this work to single NV measurements.

3.2 Level anti-crossing polarization transfer

We studied the dependence of the inhomogeneous coherence time or dephasing time T_2^*, as a function of the externally applied magnetic field, comparing between the -1 and +1 transitions. This comparison is presented below, depicting the enhanced coherence time achieved for the -1 transition near the ESLAC resonance of 50% compared to the +1 transition, indicating efficient cooling of the surrounding nuclear spin bath.
We have further measured T_2^* on NVs in an isotopically purified diamond samples, in which the concentration of nuclear spins (13C) is significantly lower. In these results we see no effect near the ESLAC, although we see some change in near the GSLAC. The underlying physical mechanisms for this effect are currently being studied theoretically.
We intend to finalize these measurements and analysis in the coming weeks and report our findings in an upcoming publication.

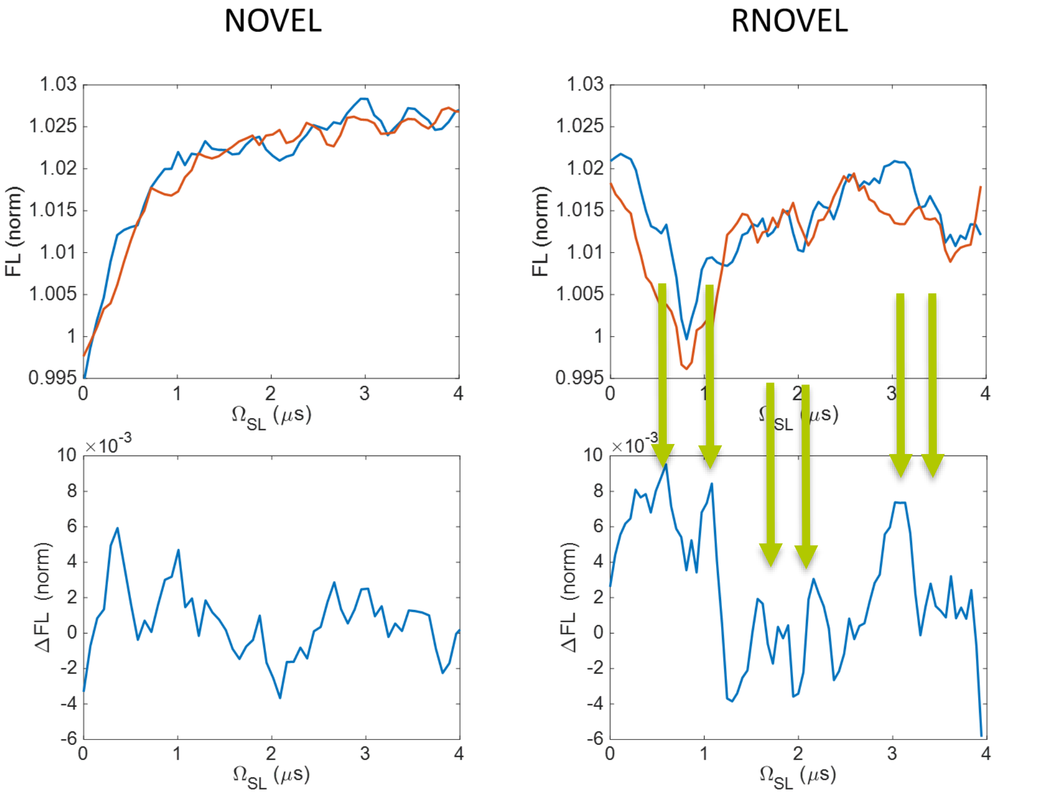

Figure 5: Comparison between NOVEL and rNOVEL, measured on an NV ensemble at a magnetic field of 350G. For rNOVEL the refocusing pulses were applied at a frequency of 0.7 MHz, and the sequences were repeated 8 times. Enhanced polarization transfer is observed for rNOVEL compared to NOVEL at the higher order resonances (marked with green arrows for clarity).

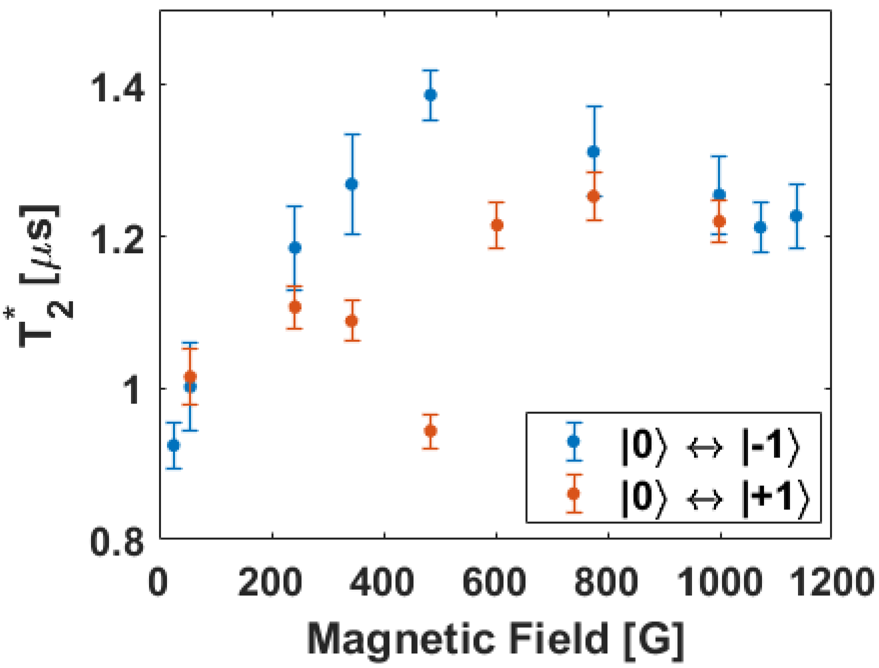

Figure 6: Measured coherence time as a function of the magnetic field, comparing between the -1 transition (blue) and the +1 transition (red). An enhancement of the coherence time by nearly 50% is observed for the -1 transition near the ESLAC (magnetic field of 512G).

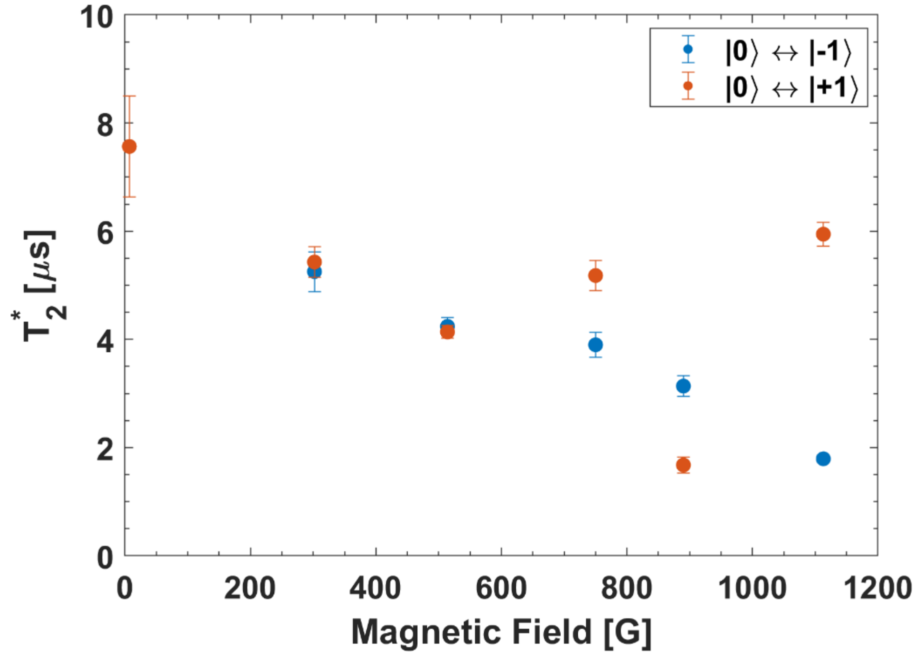

Figure 7: Measured coherence time as a function of the magnetic field, comparing between the -1 transition (blue) and the +1 transition (red), for an isotopically purified sample. Here no difference in the coherence time is observed at ESLAC, although a distinction appear near the GSLAC.

4 References

[1] D. D. B. Rao, A. Ghosh, D. Gelbwaser-Klimovsky, N. Bar-Gill, and G. Kurizki, New J. Phys. **22**, 083035 (2020).

[2] C. Belthangady, N. Bar-Gill, L. M. Pham, K. Arai, D. Le Sage, P. Cappellaro, and R. L. Walsworth, Phys. Rev. Lett. **110**, 157601 (2013).

[3] Y. Hovav, B. Naydenov, F. Jelezko, and N. Bar-Gill, Phys. Rev. Lett. **120**, 060405 (2018).

[4] K. Ben'Attar, A. Retzker, and N. Bar-Gill, In Preparation (2022).