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Optical and Spin Coherence of NV centers in Isotopically Purified Diamond for Quantum Networks

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Abstract: We discuss measurements on single NV centers in isotopically purified diamond and show coherent optical transitions combined with enhanced electron and carbon spin coherence. These results open avenues for new quantum network applications.

1. Introduction

Quantum networks could, among many other applications, enable modular quantum computation [1], which is a promising avenue in the pursuit of scalable quantum computation platforms. Defects in solids, such as the NV center in diamond, are a good candidate to realize such a network. Coherent optical transitions enable the creation of spin-photon entanglement, which can be used to connect separate nodes of the network. The NV center possesses excellent coherence and can control additional spin qubits, such as ^{13}C-spins in its vicinity, which can be used as data or memory qubits for complex algorithms. Recent progress includes entanglement experiments across three quantum network nodes [2] and fault-tolerant operations with seven qubits in one node [3].

Yet, scaling to larger networks or more complex distributed quantum algorithms remains a challenge due to the competition between the decoherence rate r_{dec} of the qubits in the nodes and the comparatively slow entanglement rate r_{ent} between nodes. To efficiently use multiple entangled states in a distributed quantum algorithm, the link efficiency $\eta = r_{ent} \,/\, r_{dec}$ is required to be much larger than unity. However, state-of-the-art experiments operate in the regime $\eta < 1$ [2].

Recent research showed that isotopically engineered diamond can host ^{13}C nuclear spins with long coherence times, in particular also during the sequences used to generate entanglement, which was projected to improve link efficiency by an order of magnitude [4]. The diamond sample of that work, however, contained a relatively high level of nitrogen impurities (~75 ppb) limiting coherence times and leading to spectral diffusion of optical transitions that hinder its use in quantum networks. It remains an open challenge to demonstrate quantum nodes in isotopically purified diamonds that combine good spin coherence with coherent optical transitions.

Here, we report an investigation of the spin and optical properties of multiple NV centers in diamonds with various ^{13}C concentrations and with low impurity concentrations. Our results reveal increased spin coherences within the isotopically purified diamond alongside low spectral diffusion of optical lines. This promising result provides a path towards demonstrations of advanced algorithms in quantum networks with link efficiencies much larger than unity.

2. Characterization of spin and optical properties

We characterize NV centers in diamonds with various ^{13}C concentrations. We consider bulk samples without structures for improved collection efficiency, such as solid-immersion lenses (SILs). The impurity concentrations are below 20 ppb for nitrogen and below 200 ppb for hydrogen, with the values set by the detection background of the secondary ion mass spectrometer (SIMS) used.

First, we measure the NV electron-spin coherence T_2^*. The electron-spin coherence also serves as an effective proxy for carbon spin coherences, with the primary distinction given by their different gyromagnetic ratios. At a concentration of 0.046% ^{13}C, we observe T_2^* values of 20-80 µs (Fig. 1a), aligning well with the anticipated range for this particular ^{13}C concentration. Notably, our findings exhibit coherence times comparable to earlier work [4] even though the present work measures NV centers at higher ^{13}C concentrations, indicating reduced impurity levels. Utilizing dynamical decoupling spectroscopy, we can also detect individual ^{13}C spins, which can be used as additional qubits [5].

At the lowest concentration, 0.002% ^{13}C, we measure coherence times of up to T_2^*=350 µs (Fig. 1b), which is lower than would be predicted from just the ^{13}C concentration. This value would be consistent with a nitrogen concentration of 20 ppb, however, we expect the measured coherence time to be limited by slow magnetic field noise. To measure longer coherence times, the magnetic field drift would need to be below 25 nT, a stringent requirement, which could be achieved by active magnetic field stabilization, not employed in this work.

Furthermore, we characterize the optical transitions of NV centers using photoluminescence excitation (PLE) spectroscopy. For this, we illuminate the sample with a green laser at 515 nm, followed by scanning a red laser at 637 nm across one of the NV center's optical transitions, while applying microwave radiation to prevent the emitter from entering a dark state.
Our initial observations reveal consistent linewidths across different NV centers. Specifically, the observed linewidths from single red-laser scans fall within a range of 50 to 80 MHz, while the combined linewidth from multiple scans of a single emitter, with a green reset pulse applied after every scan, averages around 100 MHz. The combined linewidth of 100 MHz suggests minimal spectral diffusion induced by green excitation. However, it is worth noting that the linewidth from a single scan is still considerably higher than the natural linewidth of 13 MHz [6]. We attribute this discrepancy partially to the limited photon collection without the use of a SIL. This results in relatively slow PLE scans, typically lasting around five seconds, compared to sub-second scans that can be obtained with solid immersion lenses. A more detailed investigation of these linewidths will be pursued in future work, particularly following the fabrication of SILs.

3. Conclusion

In summary, we report increased spin coherence times (T_2^*) in combination with coherent optical transitions in low-impurity diamond at different ^{13}C concentrations. These promising results can pave the way to more advanced quantum network experiments by enabling increased link efficiencies.

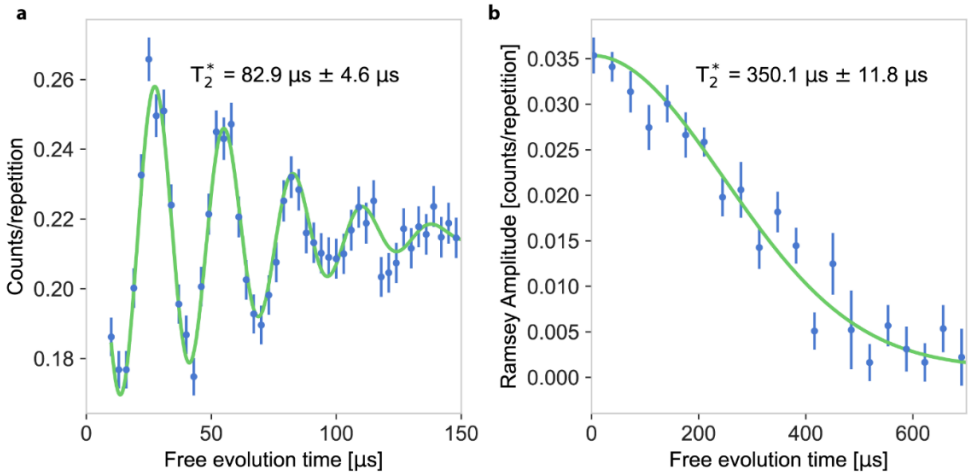

Fig. 1. Characterization of free induction decay times (T_2^*) of NV centers in an environment of (a) 0.046% ^{13}C and (b) 0.002% ^{13}C. At 0.046% ^{13}C, the measurement is performed by applying pi/2 pulses with the same phase. For the NV center at the lowest ^{13}C concentration of 0.002%, for each free evolution time separately, the coherence of the NV electron spin after the free evolution time was measured by sweeping the phase of the second pi/2 pulse, and fitting a sine wave to the measured signal. This helps mitigate the effects of slow drifts in the external magnetic field.

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