QUANTUM DEFECTS IN DIAMOND: IDENTIFYING NITROGEN ISOTOPES OF NITROGEN-VACANCY CENTERS

A PREPRINT

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ABSTRACT

This project aims to identify the isotopes present within individual nitrogen-vacancy (NV) centers in diamond. This is done to determine whether they are naturally grown ^{14}N NV centers, or if they are a product of ^{15}N ion implantation. The results are then intended to be compared to previously measured photoluminescence excitation (PLE) spectra of each NV center to study if ion implanted NV centers display greater spectral diffusion. This result impacts the reliability of the ion implantation method to produce identically emissive NV centers that can be scaled and used as reliable qubits in quantum information processing circuits.

Keywords Nitrogen-vacancy centers · qubits · quantum information · ion implantation · solid state emitters · crystal defects

1 Introduction

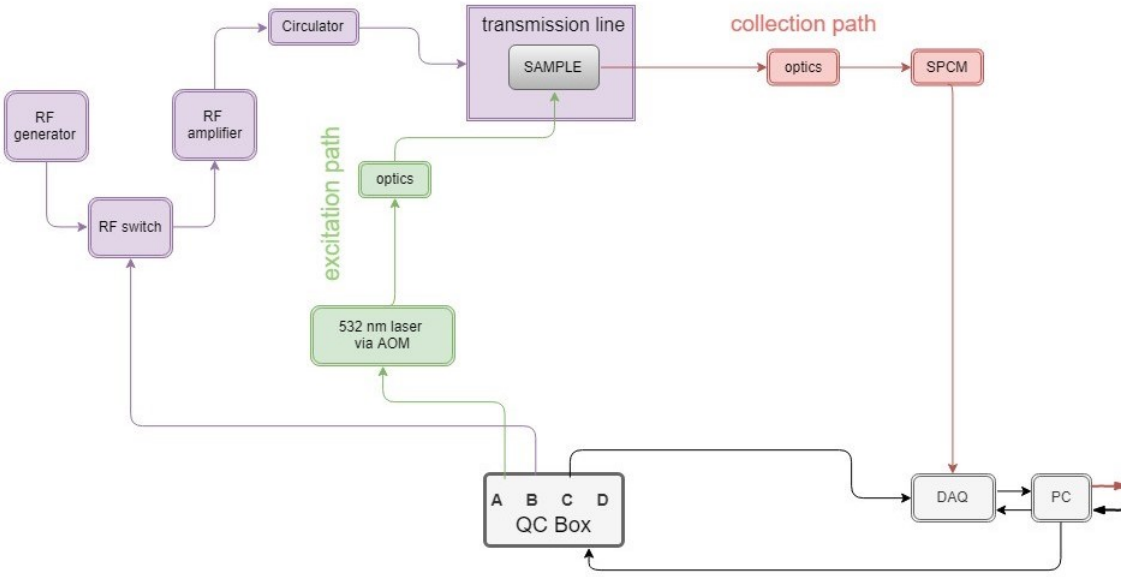
Nitrogen-vacancy centers are point defects in diamond formed by one substitutional nitrogen atom and an adjacent vacancy. This defect has been of particular interest to research in the past few decades because of its unique properties that have been recently harnessed into quantum information efforts.

Among its many unique properties is its ability to be observed optically and controlled by microwaves, while preserving quantum coherence at room temperature. This ability to resolve the spin state of a single NV center yields the ability to study the hyperfine interaction of its spin state, as well as match the number of nuclear interactions with the respective spins of the NV center's isotope.

The samples that were studied in this project were all implanted with ^{15}N nitrogen atoms. This is done by bombarding the diamond lattice with nitrogen to fill vacancies in the lattice and form NV centers. It allows for precise NV placement, which is optimal in order to ensure the homogeneity needed for scalability onto chips and photonics. However, previous studies have questioned whether this method of NV formation actually produces reliable NV centers. While ion implantation has been widely used because of the ability for precise NV placement, it may harm the NV centers by causing strain and damage to the diamond lattice, making their emission less consistent. This study aims to find whether there is a distinct correlation between the NV center's method of growth and their amount of spectral diffusion. [1]

2 Methods

In order to identify the isotope of an individual nitrogen-vacancy center, its spin state must be studied in order to resolve the hyperfine interaction of the NV center's nuclear spin states. This involves three experiments: continuous wave optically detected magnetic resonance (CW ODMR), Rabi oscillations, and pulsed ODMR. The following diagram depicts the experimental setup that was used for these tests:

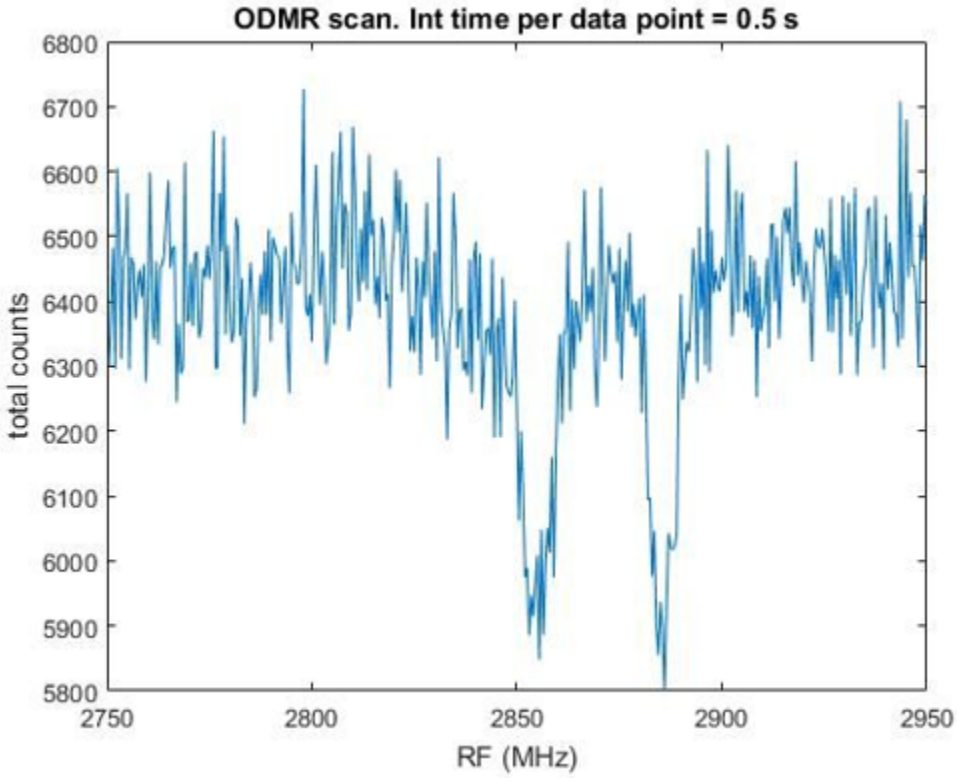


The sample is excited by a 532 nm laser, which drives the nitrogen-vacancy center's spin state to the $m_s = 0$ ground state and emits a photon ranging between 637- 850 nm. The photoluminescence of the NV center is then redirected through the collection path and the emitted photons are counted. After the spin state is initialized with the laser, the radio frequency (RF) signal is used to drive the spin state to $m_s = \pm 1$, which can be observed by a reduction in photon counts.

The experimental setup is being controlled by a Quantum Composer pulse generator box, which is controlled through MATLAB to synchronize the laser and RF pulse sequences for each experiment. The pulse generator box also triggers the timing and synchronization of the data collection process. Part of this project involved writing the MATLAB programs to run the Rabi and pulsed ODMR experiments. This was made non-trivial due to unknown limitations of the QC pulse generator box. The specific methods for each of the three experiments are described in the following sections.

3 Continuous Wave Optically Detected Magnetic Resonance

The purpose of doing continuous wave optically detected magnetic resonance (CW ODMR) scans is to determine the resonant frequency of the RF signal necessary to drive the spin state of the NV center from the $m_s = 0$ ground state to its $m_s = \pm 1$ excited state. The following figure displays a CW ODMR scan taken of a single NV center under an applied magnetic field:



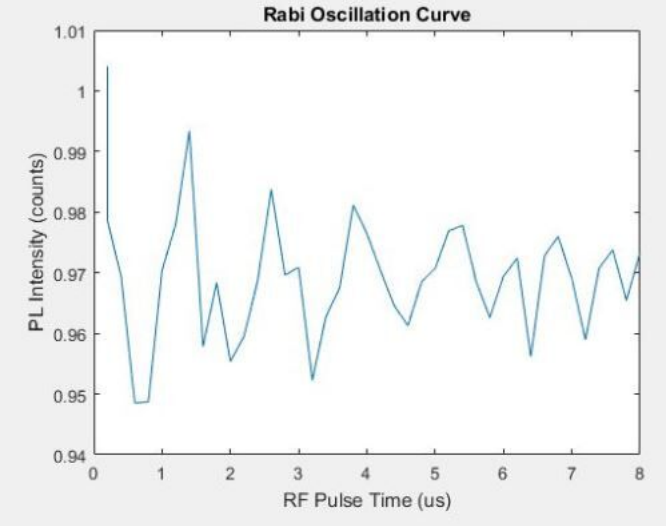
In order to separate the $m_s = -1$ and the $m_s = +1$ electron spin transitions, a weak external magnetic field is applied. From this scan, we can obtain that the energy needed to drive the NV center's spin state from the ground state to the $m_s = -1$ is approximately 2.86 GHz, and the energy to drive it to the $m_s = +1$ state is approximately 2.88 GHz. If there was no induced magnetic field, there would be no splitting of these energy levels, and the CW ODMR scan would resemble a single Lorentzian curve with a resonant frequency of 2.87 GHz.

4 Rabi Oscillation Curves

Producing Rabi oscillations by altering the decoherence rate of the NV center's spin state is vital in order to measure the π pulse time of the NV center. The π pulse time is the time it takes for the NV center's spin state to transition from its $m_s = 0$ ground state to its $m_s = \pm 1$. By varying the length of the RF pulse, the relationship between the emitted photon counts over time can be studied which results in a Rabi oscillation curve.

This is done by first initializing the NV center's spin state to zero by the laser pulse. It is then driven to the excited state by the RF pulse, at the resonant frequency that was measured from the CW ODMR. A second laser pulse is then used as a readout pulse. This compares the photon counts collected during a preset integration time, and compares these counts to the collected photon counts taken during the initialization pulse. This results in the contrast, as it subtracts the background counts from the initialization pulse. This is then used to determine how much the spin state has decayed at that specific RF pulse width. Thus, by varying the RF pulse width, it manipulates the polar angle of the spin state, and the relationship between the emitted photon counts over time can be studied.

The following figure is a Rabi oscillation that was obtained after successfully developing the MATLAB program to run the experiment:



This Rabi oscillation was taken using an ensemble NV center diamond sample because of its increased photoluminescence which made the de-bugging process simpler. Note that this oscillation does not range from 1 to zero. If there were no magnetic field induced on the sample, the maximum difference in contrast that could be observed when the population is completely transferred from its ms=0 ground state to its ms=+-1 ground state is 30 percent. Since induce a magnetic field is induced to separate the +-1 ground state, the maximum difference in PL that can be observed is reduced to 15 percent. This would translate to the Rabi curve going from 1 to 0.7 and 1 to 0.85, respectively.

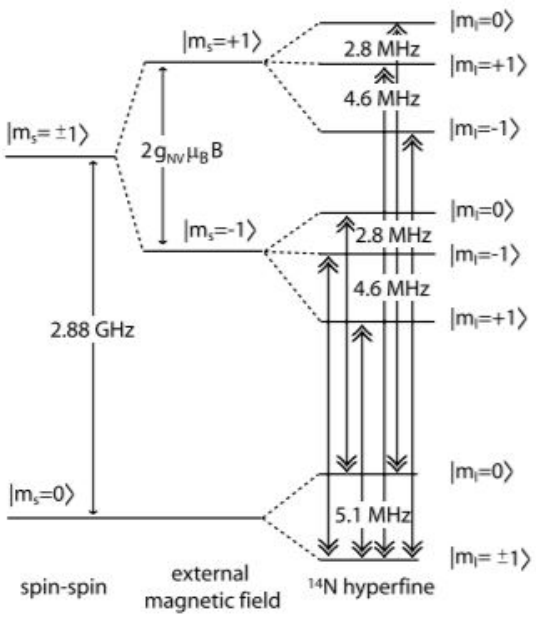
The reason why the Rabi curve initially oscillates from 1 to 0.96 (and not 0.85), is explained by the kind of sample that the measurement was taken on. That sample was an ensemble sample and not a single NV sample, so many NV centers were being excited at once instead of just one. Because an ensemble sample contains NV centers with many different orientations, they would all Zeeman shift differently because their dipole moments were pointed in different directions with the induced magnetic field. Therefore there were NV centers whose population were not driven on resonance and so their populations were not fully transferred - however they still emit optically so it dampens the signal by contributing additional light that is collected. When this test is done with the desired single NV samples, the Rabi curve is anticipated to initially oscillate from 1 to 0.85.

5 Pulsed Optically Detected Magnetic Resonance

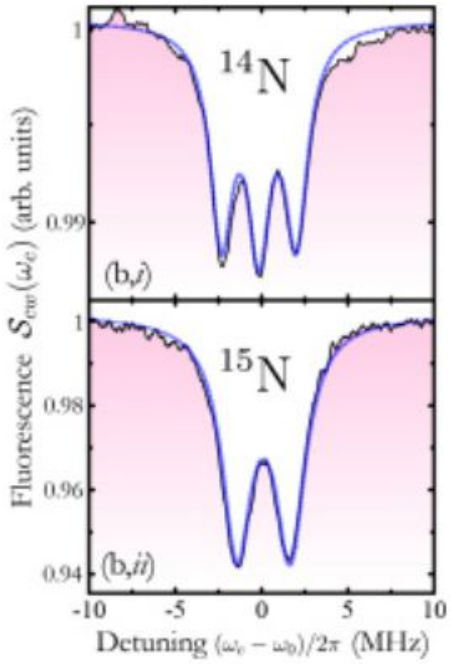
After experimentally measuring the resonant frequency and pi pulse time from the previous experiments, the final test implements these values in order to resolve the hyperfine energy levels of the NV center, and be able to identify the isotope via pulsed ODMR.

This test is essentially an adaptation of the Rabi test, with the same pulse sequence. However in this test, the length of the RF pulse width is kept constant at the π pulse time measured previously. This test varies the RF frequency,and then, by comparing the change in RF energy to the photon count, the hyperfine energy levels of the NV center's spin states can be resolved, which can be used to identify the isotope of the nitrogen.

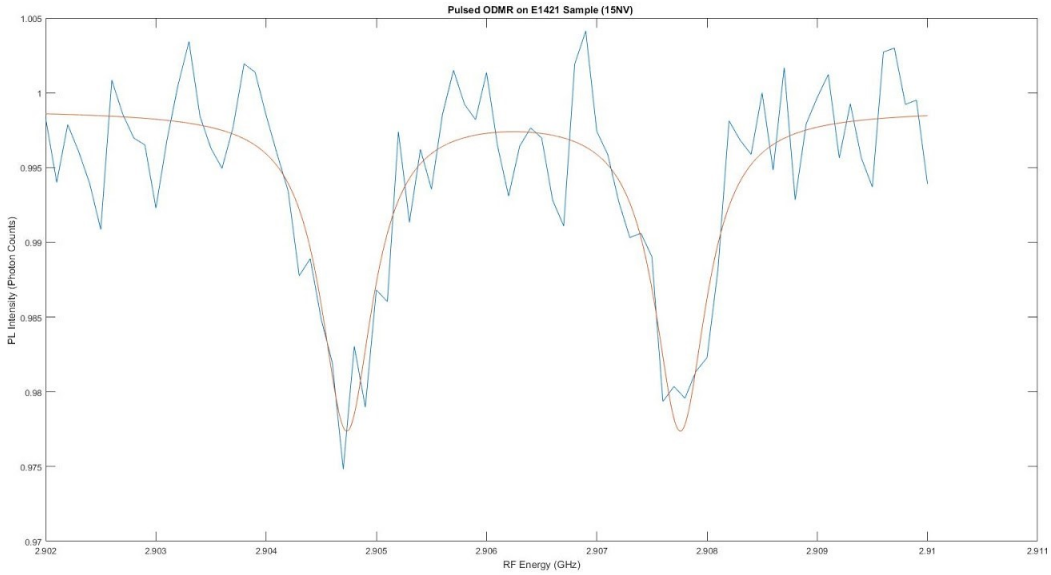
The figure below displays the energy levels of a ^{14}N NV center. [2]



It can be observed from this figure that ^{14}N, which has a spin of 1, results in three hyperfine energy levels. Similarly, ^{15}N has a spin of $1/2$, which results in two hyperfine energy levels. The resolvement of these energy levels can be observed in the following figure, where the isotope of the NV center can be identified by the number of Lorentzian dips in the pulsed ODMR scan.



This test was also tested and successfully executed on the same ensemble NV center:



It can be seen from this pulsed ODMR scan that there are two distinct dips that are separated by 30 MHz, which corresponds with what would be expected from a ^{15}N NV center, which is what this sample was implanted with. This figure is promising because it shows how the program that was developed can be effectively utilized to resolve the hyperfine structure and identify the NV center that was being studied as a $^{1}5$N NV center.

It is important to emphasize that the ability to resolve the hyperfine interactions of the NV center's spin states is not unique to pulsed ODMR: however, it is necessary in this experiment because of its ability to eliminate the effects of optical power broadening. This allows the ability to collect data much faster than if this experiment was attempted using CW ODMR, because much higher power can be used without losing a sharp line width.

If this were a Pulsed ODMR scan taken from a single NV sample, the next step would be to compare this result with the PLE taken of the same NV center to see if there is a correlation between this result and the amount of spectral diffusion that was measured for that NV center.

6 Conclusion

This project focused on developing the MATLAB programs to run the Rabi and Pulsed ODMR experiments. By collecting data using both single NV diamonds and ensemble NV diamonds, this report displayed the successful results of these experiments. In addition to developing the programs, the programs were then automated such that the pulsed ODMR tests can be run automatically for every NV center in a designated region, as well as save and document them systematically. This will increase the efficiency of the data-collecting process once the desired single NV samples are being studied. The next step of this project is to repeat these tests for the desired single NV samples that are currently being analyzed for spectral diffusion. This is done for a number of single NV centers on each sample, so the next step is to locate those exact same NV centers, repeat the tests, and then analyze to see if there is a distinct correlation between the isotope of the NV center, and how much spectral diffusion it displays.

After the isotope identifications are compared with their corresponding PLE spectra,the consequences of this possible relationship will result in a better understanding of not only whether ion implantation produces reliable NV centers, but will also result in a better understanding of how to potentially utilize naturally stable NV centers to improve emission stability of NV centers as qubits.

References

[1] S.B. van Dam, et. al. Physical Review B 99,161203 (2019)

[2] V. M. Acosta, et. al. Physical Review B 80,115202 (2009)