Understanding the Detailed Mechanism of NV Center in Diamond and Its Experimental Setup

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Abstract

In this report, we present a detailed and fundamental investigation of the mechanism underlying NV(nitrogen-vacancy) center in diamond, as well as the experimental setup required to achieve such readout. The report begins with an introduction to the electron spin, including the structure of the NV center, the energy levels of the electron, and the optical properties of the NV center. We also discuss the methods used for handling qubits, which are essential for quantum computing applications. Next, we delve into the nuclear spin and its interaction with the electron spin. We discuss the realization of quantum gates and the techniques used for manipulating the nuclear spin. We also explore the conditions required for implementing quantum non-demolition (QND) protocol, which enable single-shot readout of the nuclear spin with high accuracy and minimal disturbance. In addition to the theoretical aspects, we also describe the experimental setup required for achieving single-shot readout of the nuclear spin. Specifically, we discuss the frequency feedback control magnetic stage, which is a critical component for stabilizing the experiment and enabling accurate readout of the nuclear spin.

Keywords: *NV Center; Single-Shot Readout; QND; Magnetic Stage*

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1. Introduction

Researchers are constantly seeking good candidates for quantum computing qubits, and the NV (nitrogen-vacancy) center in diamond has emerged as a highly promising option. This defect in the diamond lattice contains two unpaired electron spins, making it suitable for use as a qubit. One of its key advantages is its long coherence time, crucial for accurate quantum computations. The NV center is also scalable, allowing it to be integrated into larger systems for more complex computations. Additionally, it operates at room temperature, simplifying experimental setup and reducing implementation costs compared to other qubit technologies like superconducting qubits that require extremely low temperatures[1].

The NV center's optical properties enable its state to be read out using the optical method (ODMR), a prerequisite for laboratory operation of the state. The nuclear spin surrounding the NV center is also a promising qubit resource due to its weak interaction with the environment and long lifetime, making it suitable for use as a quantum register[2]. However, the nuclear spin state cannot be directly read out using optical methods, necessitating indirect readout by leveraging the interaction between the electron spin of the NV center and the nuclear spin.

In the past, the state of the nuclear spin was destroyed during each single readout, which meant that the state had to be recreated before each subsequent readout[3]. This was a time-consuming process and led to reduced readout fidelity. To address this issue, we need to implement single-shot readout with quantum non-demolition (QND) protocols to preserve the state of the nuclear spin during readout[4]. The method will be introduced in detail in the context.

To improve the stability of our experiment, we have developed a magnetic stage that stabilizes the magnetic field strength based on frequency, ensuring that our experiment is conducted under a stable magnetic field and eliminating the interference from the external environment. A stable magnetic field is crucial for studying the behavior of nuclear spins and preserving their long coherence time. The present work of the magnetic stage will be exhibited in the end of the report.

2. Electron Spin

The diamond lattice is a highly symmetrical crystal structure composed of carbon atoms arranged in a face-centered cubic(fcc) lattice. One feature of diamond is the presence of point defects, which are atomic-scale irregularities in the crystal lattice. One such defect is the NV center, which stands for nitrogen-vacancy center. This defect occurs when a carbon atom in the diamond lattice is replaced by a nitrogen atom and the adjacent carbon atom is missing, forming a vacancy[5](fig. 1). The NV center forms a C_{3v} symmetry axis, which means that it has three-fold rotational symmetry around a the NV quantization axis[5](fig. 1).

2.1. Energy level

When a nitrogen atom replaces a carbon atom in the diamond lattice to form an NV center, it brings five valence electrons. Three of these electrons form covalent bonds with adjacent carbon atoms, while the remaining two form a lone pair. In

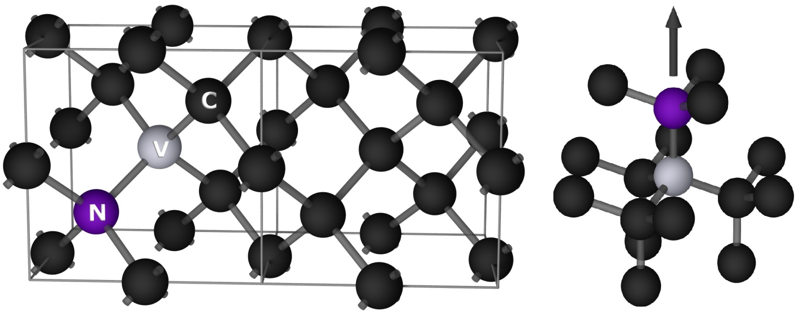

Figure 1. NV center structure. The image on the left shows two unit cells of the diamond lattice, one of which contains an NV color center. Carbon atoms are represented by dark gray spheres, while the nitrogen atom and the vacancy are marked with "N" and "V," respectively. The image on the right displays the NV center and the two shells of neighboring carbon atoms around the vacancy. The NV axis, which has a C_{3v} symmetry, is pointing upwards and is represented by a dark arrow.

addition, two of the three adjacent carbon atoms next to the vacancy will form a covalent bond between each other[2]. As a result, there is one unpaired electron in the electrically neutral NV center, denoted by NV^0. However, in most cases, we talk about the NV center with an extra electron, which is noted by NV^- (here after referred to as NV unless otherwise stated). With this extra electron, there are now two unpaired electrons in the NV center, which can form a triplet state with spin 1 or a singlet state with spin 0[2].

In the ground state of the NV center, which is represented by the $^3A_2$ and $^1A_1$ part[5] in the picture below 2, the electronic spins form three distinct energy levels in the absence of any external magnetic field. The $m_s = 0$ state in the triplet state has the lowest energy due to the pairing of electrons. In contrast, the singlet state has a higher energy level than the triplet state due to the symmetry of the special wave function, which results in a stronger electron-electron repulsion[6]. The $m_s = \pm 1$ states in the triplet state are degenerate. The energy difference between the $m_s = 0$ and $m_s = \pm 1$ states is 2.87 GHz. The singlet state is metastable,3 so it usually transits to the triplet ground state(most are $m_s = 0$), which is the reason why it is known as the intermediate state.

The triplet ground state of the NV center can be excited to the first excited state, denoted by ^3E2, by using a 532 nm green laser. The energy gap between the $m_s = 0$ and $m_s = \pm 1$ states in the ^3E state is smaller than that in the ground state, with a value of 1.42 GHz.

In the presence of a magnetic field, either from the nearby nuclear spin or an external source, the energy levels of the $m_s = \pm 1$ states in the NV center will become non-degenerate, resulting in what is known as the Zeeman effect(eZ)(see fig.

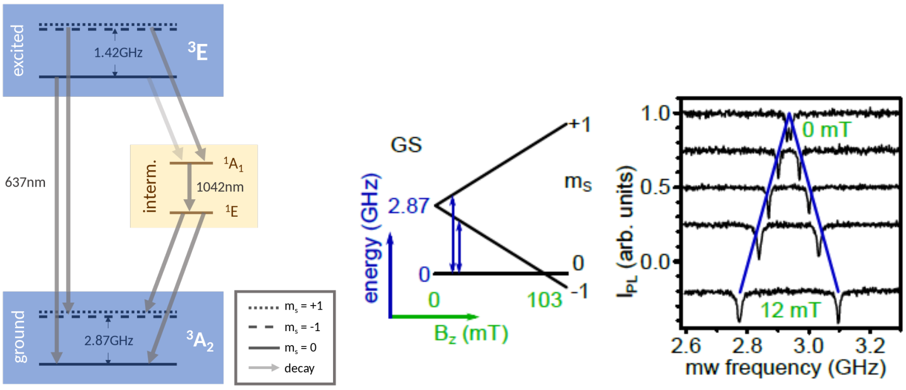

Figure 2. Left. The energy level of electron spins in NV center. The blue part is refer to the triplet state, including ground state $^3A_2$ and excited state ^3E. The yellow part is single state(intermediate state), including ground state $^1A_1$ and excited state ^1E. The grey arrows are refer to the transition process between different states[7]. **Right** Zeeman splitting of $m_s = \pm 1$ under magnetic field along NV-axis and ODMR spectrum of the $m_s = \pm 1$ state.

2 middle). The Hamiltonian of the triplet state is exhibited[2]:

$$\hat{H} = \underbrace{D_x\hat{S}_x^2 + D_y\hat{S}_y^2 + D_z\hat{S}_z^2}_{\text{ZFS}} - \underbrace{\tilde{\gamma}_e \underline{B} \cdot \underline{\hat{S}}}_{\text{eZ}}$$
$$= \begin{pmatrix} D/3 & 0 & E \\ 0 & -2D/3 & 0 \\ E & 0 & D/3 \end{pmatrix} - \tilde{\gamma}_e \begin{pmatrix} B_z & \frac{B_x - iB_y}{\sqrt{2}} & 0 \\ \frac{B_x + iB_y}{\sqrt{2}} & 0 & \frac{B_x - iB_y}{\sqrt{2}} \\ 0 & \frac{B_x + iB_y}{\sqrt{2}} & -B_z \end{pmatrix}$$

where the S is spin state of NV center and the D is zerofield splitting(ZFS) tensor, which means that the energy level still splits without external magnetic field. This is result from the magnetic dipole interaction of the two unpaired electron spins. Usually, the we aligned the magnetic field with NV axis and the E term in ZFS can be ignored, so the Hamiltonian becomes:

$$\hat{H} = D\hat{S}_z^2 - \tilde{\gamma}_e B_z \hat{S}_z$$

Due to the negative gyromagnetic ratio of electrons, the $m_s = 1$ state will have a higher energy level ($E = \gamma B\hbar/2$), while the $m_s = -1$ state will have a lower energy level, and may even have a lower energy level than the $m_s = 0$ state under a magnetic field of 103 mT. The optically detected magnetic resonance (ODMR) spectrum of the $m_s = \pm 1$ state under different magnetic field is shown in fig. 2 right.

2.2. Optical properties

In the NV center, there exist two distinct types of transitions between the excited state and the ground state, namely, radiative and non-radiative transitions[2]. The radiative transition involves the decay of the triplet excited state ^3E to the corresponding triplet ground state $^3A_2$, which results in the emission of 637 nm red fluorescence[8]. The non-radiative transition occurs when there is intersystem crossing(ISC), from the excited triplet state to the singlet state $^1A_1$, followed by the transition from the singlet state back to the triplet ground state. Typically, few photons will be emitted from the ISC process, which is difficult to detect.

Due to the higher likelihood of the $m_s = \pm 1$ state to undergo the non-radiative transition, the intensity of fluorescence emitted by the $m_s = \pm 1$ states is slightly lower (about 30% less) than that of the $m_s = 0$ state[2]. Therefore, it is possible to distinguish the electron spin states of the NV center optically, based on the fluorescence intensity.

2.3. Handle qubits

2.3.1. Initialization and Readout

The initialization of NV center will be introduced first. As previously discussed, the likelihood of the excited $m_s = \pm 1$ state to decay to the intermediate state is higher than that of the excited $m_s = 0$ state. Moreover, the intermediate state has a greater chance of decaying to the ground state $m_s = 0$[2]. Therefore, the spin state of the NV center will eventually be initialized to the $m_s = 0$ state under the continuous illumination of a 532 nm green laser.

Although the green laser can initialize all spin states to the $m_s = 0$ state, the information before the states become steady can be utilized to readout the original spin state, because the $m_s = 0$ state(red line) emits more fluorescence photons than $m_s = \pm 1$(black line)(see fig. 3). And the difference in the number of fluorescence photons emitted between the $m_s = \pm 1$ and $m_s = 0$ states is referred to as the "signal photons"(grey line).

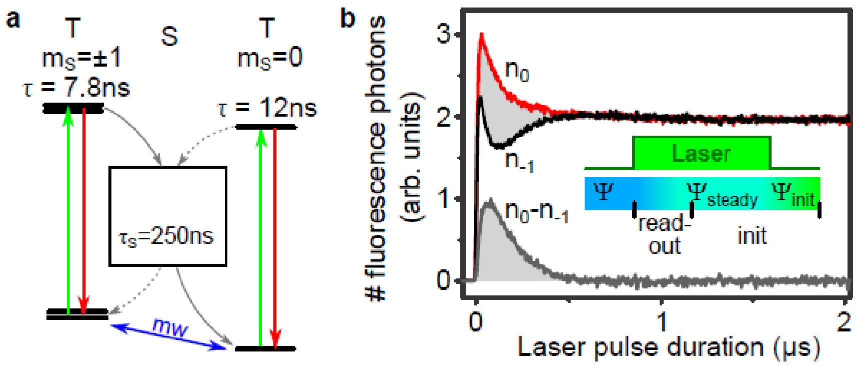

Figure 3. Initialization and Readout a shows the transition process between excited state state and ground state, attached with radative transition(red arrows), ISC(grey arrows), and lifetime of excited state and intermediate state. **b** shows the fluorescence photons of $m_s = 0$(red) and $m_s = \pm 1$(black) under laser pulse, attached with pulse sequence.

2.3.2. ODMR

Optically detected magnetic resonance (ODMR) is an optical method that can be utilized to initialize and readout the state of the NV center. There are two types of ODMR methods, continuous-wave (CW) ODMR and pulsed ODMR. The pulse sequence of CW ODMR involves the application of a continuous green laser and a mw pulse simultaneously(see fig. 4 top). The frequency of the mw is swept over time and the NV spin state is readout simultaneously. Resonance occurs when the frequency of the mw matches the energy gap between $m_s = 0$ and $m_s = -1$. However, the competition between the continuous readout laser and mw drive results in lower contrast[9]. Despite this, CW ODMR is easy to implement, and can be used to identify a rough range of the resonance peak.

To obtain a higher resolution, pulsed ODMR is developed. The sequence of this method is shown below, which begins with an initialization step, followed by a mw π pulse to flip the $m_s = 0$ state, and finally a readout pulse(see 4 top). With this sequence, the linewidth of the resonance becomes narrower compared to CW ODMR[9]. The narrower linewidth of pulsed ODMR enables the detection of narrow spectral features and provides a more precise measurement of the energy gap between the $m_s = 0$ and $m_s = -1$ states.

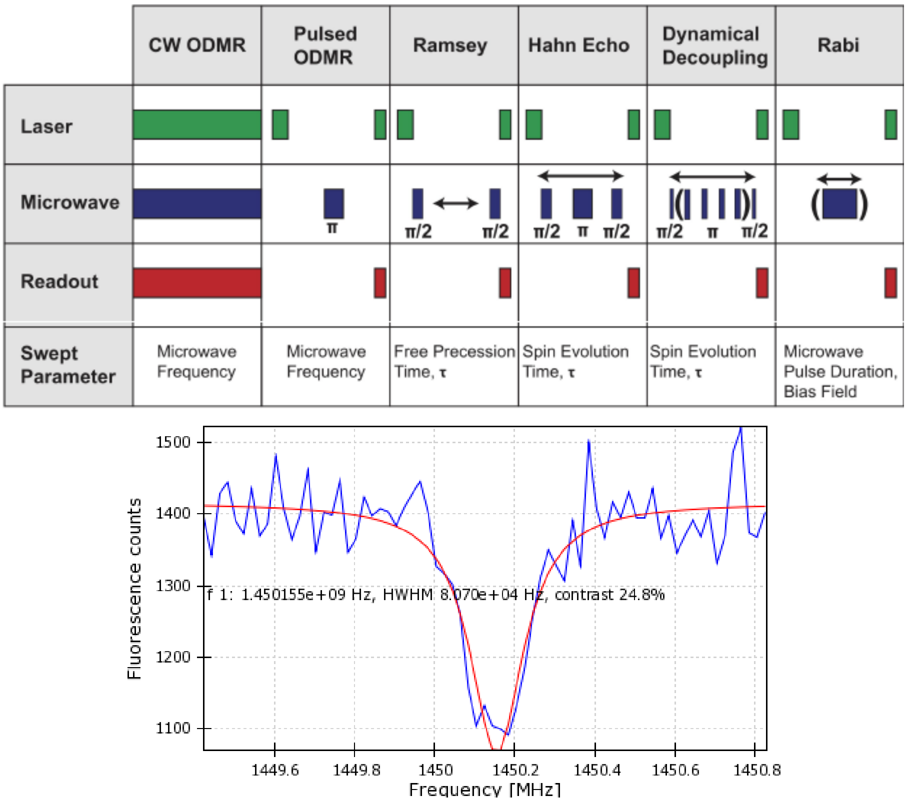

Figure 4. **Top.** The pulse sequences of several NV measurement and control methods are listed in the table[9]. **Bottom** is the pulse ODMR result in the laboratory, where blue line is raw data and red line is optimization from the blue.

2.3.3. Rabi oscillation of the electron spin

Different spin states can be generated by applying microwave (mw) pulses after initialization. The specific spin state obtained is determined by the duration of the mw pulse, which can be established by investigating the Rabi oscillation of the electron spin. The pulse sequence used for this purpose is shown on the left side of the figure 5, where the spin state is first initialized to $m_s = 0$ using a green laser pulse ($3\mu s$). After a pause of $1\mu s$, a mw pulse of length τ is applied, which causes the $m_s = 0$ state to transition to another spin state. Finally, another $3\mu s$ green laser pulse is applied to read out the state of the electron spin. The spin state can be written as

$$|\Psi\rangle = \cos\frac{\Omega\tau}{2}\,|0\rangle + e^{i\psi}\sin\frac{\Omega\tau}{2}\,|-1\rangle\,,$$

where Ω is Rabi frequency.

The result of this experiment is shown in the figure 5, where the spin state with the highest fluorescence intensity is $m_s = 0$, and the spin state with the lowest fluorescence intensity is $m_s = -1$. The intensity between $m_s = 0$ and $m_s = -1$ is corresponding to the superposition state of $m_s = 0$ and $m_s = -1$[2].

Once the Rabi frequency of the electron spin is determined, it can be used to control the spin state and perform various quantum operations, such as single qubit rotations and entanglement generation. Furthermore, the Rabi frequency is basis in different quantum gates, such as the Hadamard (H) gate or the X gate.

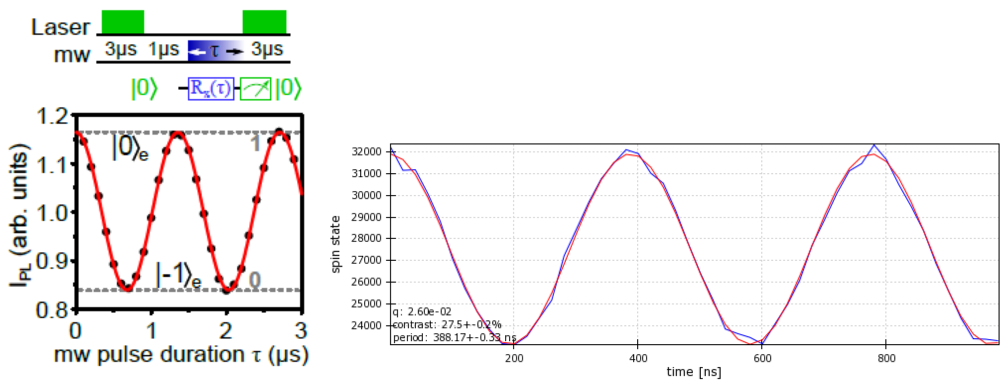

Figure 5. **Left** is relative intensity rabi oscillation of electron spin, attached with pulse sequence. **Right** is the photon counts detected of rabi oscillation in the laboratory.

2.3.4. Ramsey oscillation and Hahn Echo

Ramsey oscillation and Hahn Echo is a commonly used technique for the NV center. The Ramsey method is employed to detect the magnetic field strength along the NV axis[9], while the Hahn Echo is used to eliminate the influence on the spin state from direct current[9].

Ramsey oscillation involves applying a $\pi/2$ pulse to initialize the spin state, followed by a variable free evolution time, and then a second $\pi/2$ pulse to measure the final spin state. During the free evolution time, the spin precesses around the magnetic field direction, and the final spin state is determined by the accumulated phase shift. By varying the free evolution time, the magnetic field strength can be determined by measuring the phase shift.

In the Hahn Echo method, an additional π pulse is inserted in the middle of the free evolution time. This pulse effectively inverts the spin direction, and then re-inverts it later, resulting in a refocusing of the spin precession. This refocusing eliminates the influence of direct current on the spin state, enabling more accurate measurements of the magnetic field strength.

3. Nuclear Spin

The electron spin of the NV center interacts with several nuclear spins, including nitrogen atoms (^{15}N or ^{14}N) in the NV center, and carbon atoms (^{13}C). The weak nuclear-environment and nuclear-nuclear interaction results in the nuclear spins forming a relatively isolated spin system that is highly suitable for use as a quantum register[2]. The interaction between the nuclear spin and the electron spin can be utilized to readout the nuclear spin and to realize quantum gates such as the CNOT and the Toffoli gate.

3.1. Interaction with electron spin

The Hamiltonian governing the behavior of a nuclear spin around the NV center can be decomposed into three distinct terms:

$$\hat{H}_n = \hat{H}_{hf} + \hat{H}_{nZ} + \hat{H}_Q,$$

where $\hat{H}_{hf}$ represents the hyperfine interaction between the nuclear and electron spins, $\hat{H}_{nZ}$ is the nuclear Zeeman Hamiltonian, and $\hat{H}_Q$ represents the zero-field energy. Of these terms, the hyperfine interaction contributes most significantly to the overall behavior of the system[10].

The hyperfine interaction can be treated as an effective magnetic field B_{eff} that acts on the electron spin. Under the influence of B_{eff}, the energy levels of the electron spin triplet state will split. The nuclear spin state (m_I) determines whether the energy of the electron spin is raised or lowered. The behavior is depicted in the accompanying figure 6.

In addition to m_I, the orientation of the nuclear spin's quantization axis is also an important factor in determining the behavior of the system[2]. In the case where the quantization axis is parallel to the NV axis (case 1), such as with the nitrogen atoms in the NV center, only the transitions shown in figure 6 as black arrows are allowed, and the nuclear spin state does not change. This means that the electron spin has no influence on the nuclear spin in this configuration. In the case where the quantization axis of the nuclear spin is orthogonal to the NV axis (case 2), the hyperfine interaction is minimized and the electron spin energy levels remain unperturbed. Finally, in the general case (case 3), both "allowed" and "forbidden" transitions can occur, with the latter resulting in changes to the nuclear spin state.

3.2. CNOT gate

The CNOT gate operation involves a control qubit and a target qubit(see fig. 7 left). In the NV center, the electron spin serves as the control qubit, while the

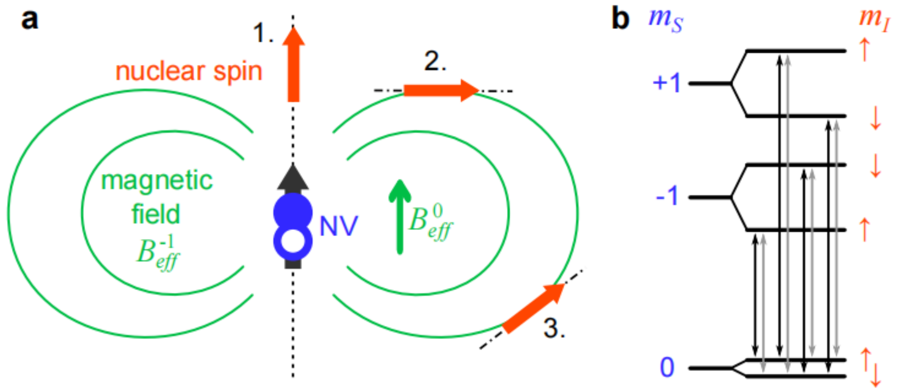

Figure 6. **a** shows the NV is under B_{eff} exerted by nuclear spin and three different orientation of nuclear quantization axis. **b** shows the energy level splitting under different nuclear spin state m_I, the "allowed"(black arrows) and "forbidden" transitions(grey arrows).

nuclear spin serves as the target qubit. To implement the CNOT gate[4], the first step of the CNOT gate operation involves initializing the electron to a known state by laser pulse and microwave. Next, a microwave pulse is applied to the electron spin, which correlates it with the nuclear spin. This correlation causes the electron spin to be in the pure state of $m_s = 0$ or $m_s = -1$ states, depending on the nuclear spin state. In addition, we also need to ensure that the state of nuclear spin is pure, since the we can not readout the superposition state of nuclear spin by electron spin. This completes the CNOT operation, correlating the electron and nuclear spins in a non-classical superposition state.

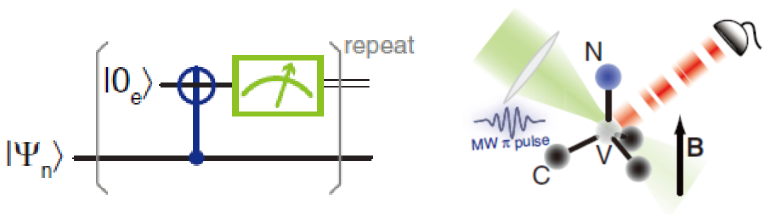

Figure 7. **Left** shows a CONT gate and $|0_e\rangle$ indicates the electron spin state $m_s = 0$ and $|\Psi_n\rangle$ indicates nuclear spin to be measured. **Right.** A naive picture of real CNOT gate implement.

4. Single-shot Readout

4.1. Methodology

To measure the nuclear spin state, the electron spin of the NV center is correlated with the nuclear spin state using a microwave pulse that is selective for the nuclear spin state. This operation is equivalent to a CNOT gate and maps a specific nuclear spin state onto the electron spin[4]. For instance, when nuclear spin is $m_I = -1$, the electron spin will flip from $m_s = 0$ to $m_s = -1$, but it does not happen when $m_I = 0$.

Then by the difference in fluorescence intensity between the $m_s = 0$ and $m_s = -1$ states, electron spin is then readout optically. Finally, we obtain the nuclear spin state according to the electron spin state.

This method destroys the electron spin state but leaves nuclear spin state undisturbed in each measurement, which highly decreases the measurement time and enhances readout fidelity. This measurement is called quantum non-demolition (QND) protocol[11].

4.2. Quantum non-demolition protocols (QND)

The implementation of quantum nondemolition (QND) protocols requires careful consideration of several conditions[4]. Firstly, there must be a measurable dependence between the nuclear spin and electron spin. This requires a sufficiently strong interaction between the two spins, which in turn requires that the distance between the nuclear spin and electron spin not be too great, and that the nuclear quantization axis not be orthogonal to the NV-axis.

Secondly, the nuclear spin state must remain unchanged under the back action of the measurement. In other words, the transition process of the electron spin must not cause the nuclear spin state to flip. This condition is satisfied, for instance, when the nuclear quantization axis is parallel to the NV-axis, as this ensures that no "forbidden" transitions occur.

Finally, the nuclear spin and electron spin must be insensitive to environmental interactions and noise. In other words, the nuclear spin should have a long lifetime and coherence time. This is necessary to ensure that the QND protocol can be implemented with high fidelity and accuracy[4].

5. Experimental setup

All experimental operations on the NV center, such as detecting fluorescence and applying microwave, radio frequency, and laser pulses, are carried out using a confocal microscope[2, 12]. The experimental setup is illustrated in the figure 8. A 532 nm laser beam is collimated through an acousto-optic modulator (AOM) and then focused onto a pinhole before being irradiated onto the diamond. The objective lens scans the focal plane to locate the NV center. A long pass filter with a 647 nm cutoff wavelength is employed to transmit fluorescence light while blocking light with shorter wavelengths, such as the excitation light. The red fluorescence is directed onto two single photon counting avalanche photodiodes (APDs) after being split by a 50/50 beam splitter. These signals are collected and analyzed by a computer, which provides feedback and control signals to the objective lens, microwave source, radio frequency source, and magnet.

The magnetic stage, driven by a servo motor, is used to maintain a stable and strong magnetic field environment. To ensure stability, we dynamically modulate the magnetic field strength using the resonance frequency from ODMR method. We have completed the program for automated detection, and the full functionality will be implemented in subsequent work.

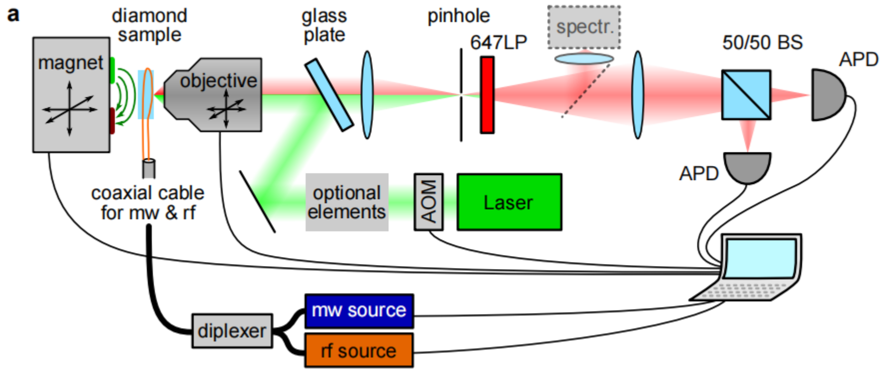

Figure 8. The experiment setup of confocal microscope

6. Conclusion

In conclusion, the NV center in diamond and its surrounding nuclear spin hold great promise as qubits for quantum computing. The NV center's desirable spin and optical properties, including high sensitivity to magnetic fields, and efficient optical initialization and readout. Additionally, the nuclear spin's weak interaction with the environment and long lifetime make it a promising resource for use as a quantum register. To utilize the NV center and its surrounding nuclear spin as qubits, various quantum protocols can be employed, such as ODMR and Rabi oscillation. Single-shot readout techniques can be used to efficiently and non-destructively obtain the nuclear spin state. Furthermore, experimental techniques such as the confocal microscope and magnetic stage have been developed to achieve qubit operations and high-precision and stable measurements.

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