Graduate School of Pure and Applied Sciences

Measurement systems for the control of the electron spins in nitrogen-vacancy centers in diamond at room and low temperatures

(ダイヤモンド窒素-空孔中心電子スピン制御のための室温及び低温における測定システム)

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Abstract

1. Introduction

Nitrogen-vacancy (NV) centers in diamond are point defects in the diamond lattice constituted by a nitrogen atom and an adjacent vacancy. NV centers are constituted by a ground state spin triplet (S = 1, m_s = 0, -1, +1) with a transition at a MW frequency of 2.87 GHz, which is referred to zero-field splitting (ZFS). This transition is sensitive to environmental conditions such as AC and DC magnetic fields, strain and temperature [1]. Since the electron spin of NV centers can be initialized and manipulated at room temperature and it has a long coherence in the order of milliseconds [2], NV centers have become an attractive platform for sensing and quantum information.

Another advantage of NV centers is the possibility to implement polarization, manipulation, and detection of the spin with relatively simple measurement setups. The spin can be optically initialized on a timescale of a few microseconds by using laser pulses from a laser diode at a wavelength of 520 nm. By applying MW pulses with a wire near the diamond surface, the spin can be manipulated and flipped between the transitions of the ground state. Upon laser illumination, NV centers emit photoluminescence in a range of 600-800 nm. Thus, the electron spin resonance transitions can be optically measured, giving the so called optically-detected magnetic resonance (ODMR). The photoluminescence intensity reflects the spin polarization, thus making it possible to probe the spin dynamics by measuring the change of emitted light with photodiodes or CCD/CMOS cameras.

Sensing with NV centers is strongly influenced by the distance between them and the sample to be sensed. In synthetic diamond substrates fabricated with chemical-vapor deposition (CVD), NV centers are randomly distributed in the volume during substrate growth. To take advantage of the sensing capabilities of NV centers, the density and spatial location of the defect can be artificially controlled by using techniques of ion implantation [3]. For instance, a shallow ensemble of NV centers implanted in a 2D layer at a distance < 100 nm from the surface of the diamond was found to be suitable for imaging magnetic samples with a spatial resolution of about 500 nm. Each NV center of the ensemble behaves as a single nano-detector which senses a point on the sample's surface placed in contact with diamond; for instance, imaging the magnetic field distribution over an electric circuitry. By acquiring the photoluminescence of the NV center ensemble with a camera, a spatial characterization of the sample becomes possible [4].

Despite the well consolidated technology to measure NV centers there are still technical challenges that have to be addressed to measure the electron spin state at room and low temperature. An ensemble of NV centers is useful to increase the signal-noise ratio of the measurement, proportional to $N^{1/2}$, where N is the number of driven spins. However, to benefit from the NV ensemble, each spin has to be manipulated in an identical way. For instance, field gradients and spatial inhomogeneities of the MW driving source cause a degradation of the signal due to the average of the ensemble. On the one hand, different techniques have been employed to generate uniform MW fields across the diamond sample, such as planar loop gap resonators or 3D dielectric resonators which use large current loops. On the other hand, an increase of the field uniformity implies an excitation field with low intensity, which limits the speed of the spin manipulation and imposes the use of large MW powers. The trade-off between uniformity and field intensity leads to a search for more efficient ways to manipulate the spin and maximize the Rabi frequency gain. The Rabi frequency gain is defined as the Rabi frequency of the NV center's electron spin, measured per square root of the MW power (with units $MHz/W^{1/2}$) delivered to the device used the excitation. Low power operation is important to realize diamond-based integrated devices, to reduce the electric noise and avoid the heating of the samples to be sensed such as biological samples. At low temperature, high MW powers introduce thermal heating in the cryostat which is not beneficial for a stable operation. Low temperature setups are often used to characterize superconductive thin film with NV centers and high MW powers may interfere with the superconductive state.

In this research, three types of optical setups to measure the ensemble of NV centers at room and low temperature are designed and constructed. The main purpose of the research is to address technical challenges about the MW excitation of the electron spin of NV centers in measurement setups with different size, detection system, and environmental conditions. The first setup is constituted by a portable device based on a microcontroller, the second one by a tabletop setup with imaging capabilities, and the third one by a closed-cycle cryostat for ODMR measurements at low temperature. Efficient MW antenna or resonators are designed to specifically operate in each setup, at a low MW power consumption, and at the same time to deliver high intensity MW magnetic fields. The MW fields of the devices are characterized by measuring the electron spin of NV centers. Strategies of design and procedures of construction of the setups are reported, showing different methods for the optical excitation and detection of the electron spin.

2. A portable measurement setup based on microcontroller

The first part of the research focuses on the development of a portable measurement setup based on a microcontroller to performed experiments of pulsed magnetic resonance in the diamond. The system is fully implemented by using an Arduino Uno board equipped with an AVR microcontroller which is used as a TTL pulse sequencer to drive precise laser and MW pulses with a resolution of 62.5 ns. The whole equipment is assembled with low-cost modules on printed circuit boards (PCB) and placed in a compact box with a volume of 20 x 40 x10 cm^3 and a weight of 1.8 kg. A compact detection system based on a switched integrator and a photodiode in the vicinity of a diamond substrate is used to measure the photoluminescence from NV centers (Fig. 1). The output signal is directly read by the microcontroller, oversampling the analog-to-digital converter (ADC) of Arduino Uno. We characterize a CVD diamond sample by performing the pulsed optically-detected magnetic resonance (ODMR). A planar MW antenna on PCB is designed and constructed to efficiently drive the [111] transition of the electron spin ensemble in an area of 1 mm^2. By using the MW antenna, a coherent manipulation of the electron spin of NV centers by driving Rabi oscillations with a Rabi frequency gain of about 7 $MHz/W^{1/2}$ is demonstrated. This gain makes it possible to operate within the bandwidth of 8 MHz of Arduino Uno, at MW powers below 1 W. We demonstrate different pulse sequences to study electron spin relaxation and dephasing. An antenna to simultaneously deliver MW and radio-frequency (RF) pulses is developed to extend the number of possible experiments by the multifrequency manipulation of the electron spin. A Rabi frequency gain of about 14 $MHz/W^{1/2}$ is obtained at MW frequencies and a gain of 4 $MHz/W^{1/2}$ is obtained at RF frequencies. By using the multifrequency excitation, dressed states of the electron spin are created, which are used to extend the spin nutation over 2000 μs, characterize RF fields, and generate new levels with sensing capabilities. Compared to the previous works [5], our system results in a versatile setup with significantly reduced complexity, taking advantage of the open-source environment of Arduino Uno. The emerging field of quantum information requires specialized technical courses to instruct the new generations about the concepts of quantum bits and spin manipulation. Then, the portable setup finds application as a learning module in classrooms of academia and in research laboratories and enables a wider audience to access the magnetic resonance in diamond.

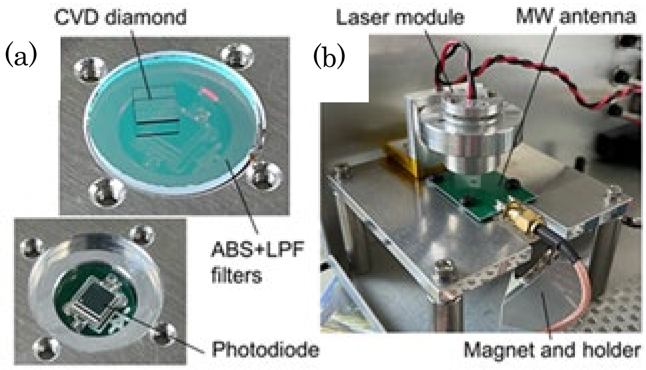

Fig. 1 (a) Detection system of the portable setup. (b) Arrangement of the sample holder.

3. A table-top setup for wide-field imaging

In the second part of the research, MW antenna and resonators with a large Rabi frequency gain are designed and constructed. The characterization of these structures is performed by using a table-top setup with widefield imaging capabilities and a diamond sample with shallow implanted NV centers at a distance of 10 nm from the surface of the diamond. The key point to obtain a large Rabi frequency gain was found to be using a magnetic field of a structure resonating at MW frequency, with a MW current confined in constrictions with the size comparable with the skin depth (~2 μm) and by probing the magnetic field at a distance of few micrometers by using shallow implanted NV centers.

First a system for the remote control of the electron spin by using exchangeable resonators is developed [6]. In this system, the main excitation source constituted by a planar MW antenna on a PCB is used to couple patterned Au resonators on silicon substrates. The diamond is sandwiched between the two structures with the NV layer in contact with the surface of the Au resonators. A distance of about 0.5 mm exists between the MW antenna and the coupled resonators. The planar MW antenna creates a moderate but homogeneous magnetic field across the sample in an area of 1 mm^2. The Au resonator on silicon produces a localized and enhanced magnetic field in micrometer-sized constrictions placed at the center of the resonator (Fig. 2a). An enhancement of 22 times for the magnetic field is obtained on the Au resonators, with a Rabi frequency gain of about 154 $MHz/W^{1/2}$.

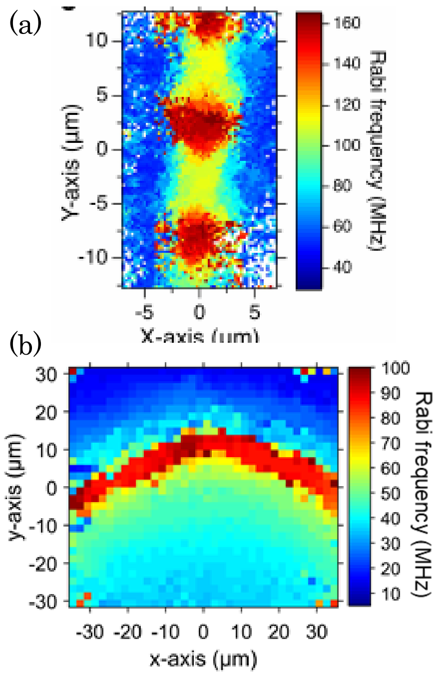

Fig. 2 Imaging of the Rabi frequency distribution. (a) Constriction of an exchangeable resonator for a MW power of 35 dBm. (b) Edge of a drop-shaped antenna at a MW power of 21.8 dBm.

To obtain a higher gain, another system based on a planar MW antenna on a PCB is developed. The design consists of a drop-shaped loop-gap antenna with a pure capacitive gap and a minimum loop size of 0.13 mm, which is used to increase the magnetic field within the loop and obtain the impedance match with the 50 Ohm feed line. By polishing the surface of the antenna and probing the confined MW current of the resonance mode by using shallow implanted NV centers (Fig. 2b), a Rabi frequency gain of 257 $MHz/W^{1/2}$ is obtained for a magnetic field localized to a 5 μm current sheet.

4. A setup to perform ODMR measurements of NV centers at low temperature

In the third part of the research, a setup to perform ODMR measurements of NV centers at a low temperature is developed. At a low temperature, NV centers have been used to characterize vortices and the Meissner screening in type II superconductive thin films [7]. Cryostats with small chambers operating in a liquid helium bath are typically employed. Small chambers can not host optical elements, and thus objective lenses with a long working distance (~10 mm) and a low collection efficiency have to be placed outside the cryostat. Low laser and MW powers are also required to reduce the heat flow to the samples. With these limitations, small contrasts in the photoluminescence of NV centers require long measurement times to obtain a good signal-noise ratio. The bath of liquid helium has the benefit to reduce the vibrations of the cryostat when atomic force microscope (AFM) probes are used but it limits the operation to a few days. Then, a close-cycle cryostat with a large chamber capable to host optics and with long operability is preferred.

In this research, an optical setup with capabilities of widefield imaging and a 4 K closed-cycle cryostat is used for ODMR measurements of an ensemble of NV centers at a low temperature. The sample is installed on a piezoelectric stage and the position is controlled with a spatial resolution of 500 nm, with an estimated imaging resolution of about 700 nm. 3D superconductive Helmholtz coils made of a NbTi wire were designed and constructed to produce the static magnetic field to control the Zeeman splitting of the NV center's electron spin (Fig. 3a). The magnetic field of the coils was characterized by using NV centers. The reduction of the heat flow towards the 4 K stage of the cryostat is analyzed and suitable materials to realize thermal anchors and current leads are chosen. The superconductive coils operate with currents within 1 A and the generated magnetic field at the sample position was characterized by measuring the ODMR of NV centers. An excellent agreement was found between the theoretical and experimental results. Different types of MW excitation sources are adapted for use at a low temperature, such as a patterned Au resonator on silicon substrate or an antenna on a PCB. An antenna with a rectangular hole to host samples is proposed to study the Meissner screening of superconductive materials.

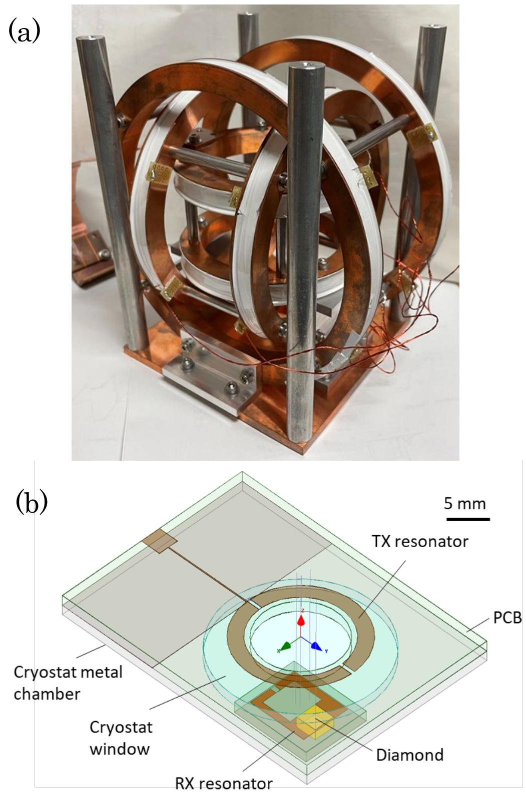

Fig. 3 (a) 3D Superconductive Helmholtz coils. (b) System for the wireless MW excitation in small cryostats.

Small cryostats such as the Janis ST-500 are often used for the PL spectroscopy of NV centers at low temperatures. However, this type of cryostat has a small chamber ~ 2.5 cm^3, for which the heat inflow has to be limited for a stable operation. The limited size of the chamber does not facilitate the electrical connections and a small and thin wire is used for the MW excitation, limiting the area of measurement near the wire. The preparation of the wire is also time-consuming and the position can not be changed after closing the cryostat. A system of wireless MW excitation is developed to excite NV centers from outside the cryostat (Fig. 3b). The system is based on the resonant inductive coupling between a transmitter antenna placed outside the cryostat and a receiver loop resonator placed inside the cryostat. The receiver is constituted by a large loop which can host the diamond sample. ODMR measurements of NV centers were successfully obtained by using a wireless MW excitation at a distance of 5-12 mm from the diamond sample. This system finds its application in cryostats with small chambers and to excite NV centers in diamond samples with electrical devices or electrodes patterned on the surface used to control the charge state of NV centers.

5. Conclusions

In this research, experimental setups to measure NV centers in diamond at room and low temperature were developed. In the first part, a portable setup was assembled in a compact box of 20 x 40 x 10 cm^3, with a lightweight of 1.8 kg. The coherent manipulation of the NV center's electron spin was demonstrated by using TTL pulses of a microcontroller, with a resolution of 62.5 ns. Rabi oscillations with a frequency up to 6 MHz were measured with a MW power below 1 W. In the second part of the research, a tabletop setup for the wide-field imaging of localized MW magnetic fields by using shallow implanted NV centers was constructed. A system for the remote control of the electron spin by using exchangeable resonators was developed. An enhancement of 22 times of the MW field measured on micrometer-sized constrictions of the resonators was obtained, as compared to the bulk Rabi oscillations generated by using a planar ring MW antenna in an area of 1 mm^2. A new design of a drop-shaped antenna on a printed circuit board was fabricated to achieve a large Rabi frequency gain of 257 $MHz/W^{1/2}$, confined in a 5 μm current sheet. In the third part of the research, a setup for the ODMR measurements of NV centers at low temperatures with a closed-cycle cryostat was constructed. Superconductive Helmholtz coils were designed, fabricated, and characterized by using the ODMR spectrum of NV centers. To control the frequency of a single resonance transition of NV centers, a gain of 0.188 MHz/mA was obtained experimentally, in excellent agreement with the theoretical calculation. Finally, a system for the wireless excitation of the electron spin from outside a cryostat with a small chamber (~ 2.5 cm^3) was developed. The ODMR of NV centers was successfully measured by excitation at a distance in a range of 5-12 mm, by using the resonance inductive coupling between two resonators placed outside and inside the cryostat.

Efficient MW antenna or resonators with high gain and operating at low powers are crucial in several situations to avoid the heating of samples or reduce the heat flow at a low temperature. Moreover, the resonance transitions of NV centers are temperature dependent with a fluctuation of about 84 kHz/K at room temperature, which is important to maintain stable environmental conditions when applied to sensing. Low radio-frequency powers are preferred in integrated and compact devices, where the MW fields may interfere with nearby devices. In quantum information processing, high gains are required to achieve a spin manipulation faster than its decoherence rate and thus to avoid potential failures in quantum protocols.

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