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Non-intrusive tunable resonant microwave cavity for optical detected magnetic resonance of NV centres in nanodiamonds

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ABSTRACT

Optically detected magnetic resonance (ODMR) in nanodiamond nitrogen-vacancy (NV) centres is usually achieved by applying a microwave field delivered by micron-size wires, strips or antennas directly positioned in very close proximity ($\sim \mu\text{m}$) of the nanodiamond crystals. The microwave field couples evanescently with the ground state spin transition of the NV centre (2.87 GHz at zero magnetic field), which results in a reduction of the centre photoluminescence. We propose an alternative approach based on the construction of a dielectric resonator. We show that such a resonator allows for the efficient detection of NV spins in nanodiamonds without the constraints associated to the laborious positioning of the microwave antenna next to the nanodiamonds, providing therefore improved flexibility. The resonator is based on a tunable Transverse Electric Mode in a dielectric-loaded cavity, and we demonstrate that the resonator can detect single NV centre spins in nanodiamonds using less microwave power than alternative techniques in a non-intrusive manner. This method can achieve higher precision measurement of ODMR of paramagnetic defects spin transition in the micro to millimetre-wave frequency domain. Our approach would permit the tracking of NV centres in biological solutions rather than simply on the surface, which is desirable in light of the recently proposed applications of using nanodiamonds containing NV centres for spin labelling in biological systems with single spin and single particle resolution.

Keywords: microwave cavity, dielectric loaded cavity, nanodiamonds, ODMR, ESR

1. INTRODUCTION

Over the past few decades a diamond defect, known as the nitrogen–vacancy (NV) centre, has attracted great interest. The diamond NV centre possesses an atomic-scale electronic spin state that can be used as an individually addressable solid-state quantum bit (qubit) with an extremely long coherence time ($\sim \text{ms}$)⁵ even at room temperature³⁻⁶. The NV centre shows a zero phonon line at 638 nm with phononic side bands resulting in a broadband emission from 650 to 750 nm. Recent experiments have shown that the NV centre can be used as a low magnetic field gradient sensor and temperature sensor⁷⁻¹¹. Diamond is an inert material with a limited photo-blinking, which enables bio-cell imaging and bio-molecular tracking due to the absence of photo-bleaching¹²⁻¹⁴. Electron spin resonance (ESR) is typically used for identifying the NV centre in diamond even when embedded in biological systems¹⁵⁻¹⁶. The excitation of the NV centre with a microwave field resonant with the zero field splitting of the ground state ($m_s=0 \leftrightarrow m_s=\pm 1$) and with an external magnetic field, alters, respectively, its steady state population and the separation in energy between the otherwise degenerate sublevels $m_s=+1$ and -1 . A corresponding change in the fluorescence of the centre is observed and its detection can be used to measure directly the distribution of the local magnetic field with, in principle, single spin resolution¹⁷⁻¹⁸.

2. ODMR: OPTICALLY DETECTED MAGNETIC RESONANCE

Optically detected magnetic resonance (ODMR) differs from electron spin resonance (ESR) as in the former the transitions between spin sublevels are detected optically rather than by absorption or reflection of microwaves as per the

latter. A significant advantage of using ODMR for NV centres in diamond is that it does not rely on extremely low temperatures (\sim K) or high magnetic fields (\sim 10 T) to create spin polarization from the thermal Boltzmann distribution. Optical pumping of the NV with the excitation/readout laser naturally causes a non-Boltzmann steady-state spin alignment of the centre in the ground state at room temperature and zero magnetic field. Diamond NV centres feature 0.7 quantum efficiency¹⁹. They possess a homogeneous optical spectral broadening line width as small as 50 MHz and an electron spin decoherence time exceeding 50 μ s at room temperature¹. In the NV, a resonant microwave field (\sim 2.88 GHz) can drive the transition between the $m_s = 0$ and $m_s = \pm 1$ ground spin sublevels. The fraction of the population that is driven to the $m_s = \pm 1$ state, once optically excited to the electronic excited state, can decay non-radiatively without emitting a photon via a metastable state. This results in a decrease in fluorescence intensity from the NV centre. Practically, when the NV centre is pumped optically (for instance with an off-resonance laser at 532 nm) and its emission is measured, a microwave field can be applied and swept in frequency, when the microwave frequency becomes resonant with the transition ($m_s = 0 \leftrightarrow m_s = \pm 1$) a decrease in the fluorescence intensity is observed. Traditional techniques to measure ODMR in diamond NV centres use coplanar waveguides or, more crudely, thin wire antennas suspended over the nanodiamonds to carry the microwave field. These methods are however usually quite inefficient. They require a lot of power, which undesirably produces local heating. Also, they do not produce homogeneously distributed spatial fields and, as a result, they require the nanodiamonds to be located in close proximity (\sim μ m) to the structures acting as microwave antennas. Figure 1 shows the energy level diagram of the NV centre.

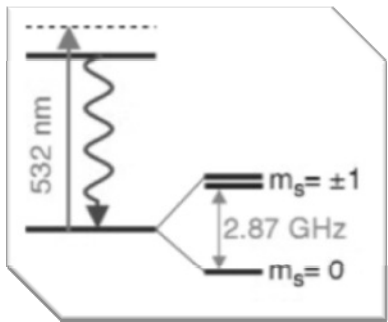


Figure 1: Schematic, simplified representation of the energy level scheme of the diamond NV centre. The triplet ground and excited states are shown as well as the $m_s = 0$ and the $m_s = \pm 1$ ground-state spin sublevels.

In this paper, we propose an alternative method to deliver the microwave field to the NV centre when performing ODMR measurements. It exploits the use of a custom-built resonator. Such a resonator offers two main advantages over the aforementioned standard methods. Firstly, it is more flexible as it is mounted over the nanodiamond sample and can be freely moved in space with respect to it. Secondly, it produces a more homogeneous microwave field with relatively less power. The basic concept is shown in figure 2.

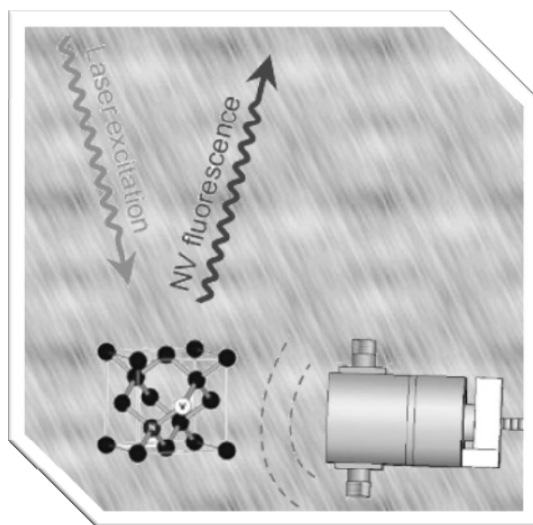


Figure 2: Schematic representation of the ODMR measurement. A laser beam shines nanodiamonds hosting NV centres, while a microwave field produced by our custom-made resonator is used to induce the decrease in fluorescence.

3. ELECTROMAGNETIC RESONANCES IN MICROWAVE CAVITY

Dielectric Resonators are very useful in many industries that require radar detection, proximity detection, as well as military based applications like secure transmissions, remote guiding, navigation and positioning systems²⁰⁻²¹. In the past, the use of whispering gallery mode resonators has proved to be a very accurate method for measurements of the complex permittivity of extremely low-loss dielectrics²². Thus, ultra-sensitive transducers can be developed by converting the particular quantity of interest into a frequency measurement. The method has been employed for very precise measurements of the permittivity and the dielectric losses of both isotropic and uniaxial anisotropic materials as well as the determination of the susceptibility added by paramagnetic impurity ions. For example, the chromium Cr^{3+} ion present in Sapphire (Ruby) introduces a well-described electron spin resonance (ESR) that has been exploited as a ruby maser²³⁻²⁷. Depending on the application, the requirement on material properties and size of the dielectric, the make-up of the resonator can vary substantially. To make the right choice of material and dimensions it is very important to use precise experimental and numerical techniques²⁸⁻³⁴ to properly characterize materials. In this paper we present the design of the cavity based on rutile, we use for this experiment.

Two types of electromagnetic modes that exist in cylindrical systems are commonly used to build dielectric resonators for generating a signal source, or a band-pass filter, Whispering Gallery (WG) modes (in general hybrid electromagnetic modes) and pure Transverse modes. WG modes in a cylindrical resonator have a particular feature, which confines the energy into the dielectric resonator. However, there is a high density of spurious modes, which is the reason why in some cases we use an open cavity³⁵⁻³⁶. On the other hand, Transverse modes have an azimuthal mode number of $m=0$ and there are only 3 field components. Structures using these conventional modes offer a better frequency mode isolation than the whispering gallery modes (i.e., smaller spurious mode density). Nevertheless their quality factor is often limited by the metallic wall losses due to lower confinement when compared to WG modes. If the metal losses induced by the penetration of the tangential magnetic field components of the mode are a problem, it is possible to plate the internal wall of the cavity with silver (to reduce the surface resistance) and even make it look like a mirror. This will increase the quality factor of the cavity.

For this experiment, we need to build a cavity that resonates at 2.87GHz and where the field should be radiated over a sample size of about 10mm x 10mm. To achieve this we built a hollow high-permittivity dielectric ring loaded in a cavity with no bottom lid. The high permittivity reduces the size of the cavity proportionally to $\epsilon_r^{1/2}$. Each NV has a unique resonance frequency, thus it is necessary to tune the resonance frequency of the cavity. The transverse electric mode can be tuned by inserting a metal plunger that disturbs the mode propagation and results in a resonance frequency shift. To compare both the coplanar and the microwave cavity techniques, we chose the electromagnetic mode with the

same field propagation directions over the sample. The adjusted resonance frequency will help to set the specific frequency of the NV centre, which is optically pumped.

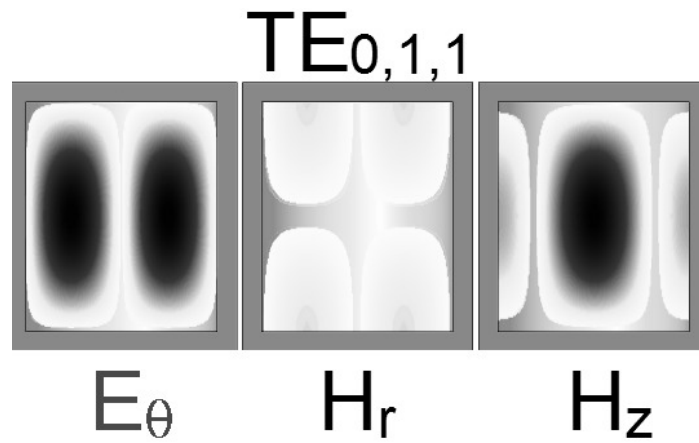


Figure 3: Illustration of the absolute field density of the transverse electric mode TE_{013} with zero azimuthal mode number and one radial and axial variations.

The cavity is designed to work on a transverse electric (TE) mode that will be adjustable from 2.7 to 3.1GHz corresponding to the largest frequency separation of the transitions $m_s = \pm 1$ under applied DC magnetic field. High permittivity low loss microwave materials for confining the field over a 10x10mm sample are not numerous. TiO_2 is one of them⁴, however the fundamental mode ($TE_{0,1,1}$) in the cavity is very low in frequency, 2.2GHz, therefore we need a higher order of mode such as the $TE_{0,1,3}$ mode (one radial and three axial variations) resonating at 2.7GHz. The insertion of the plunger will increase the $TE_{0,1,3}$ resonance frequency up to 3.1GHz.

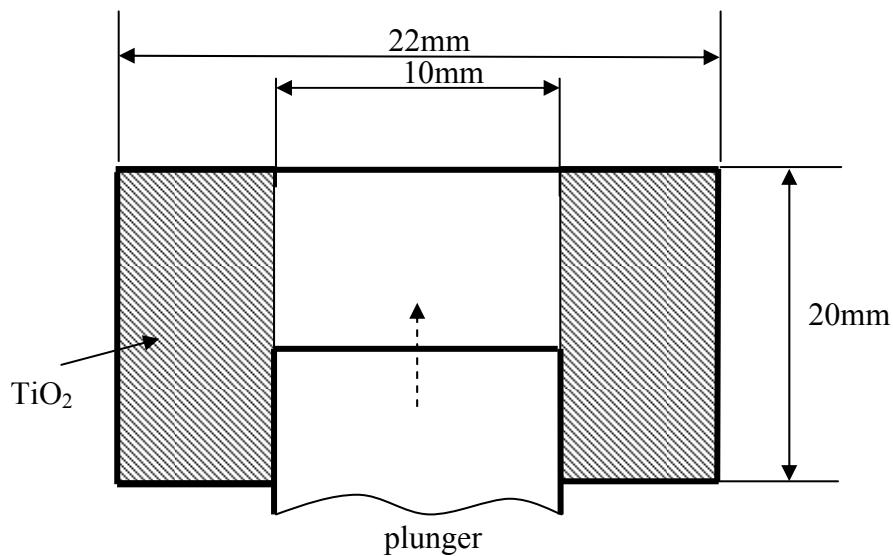


Figure 4: Schematic of the mechanical design of the microwave cavity

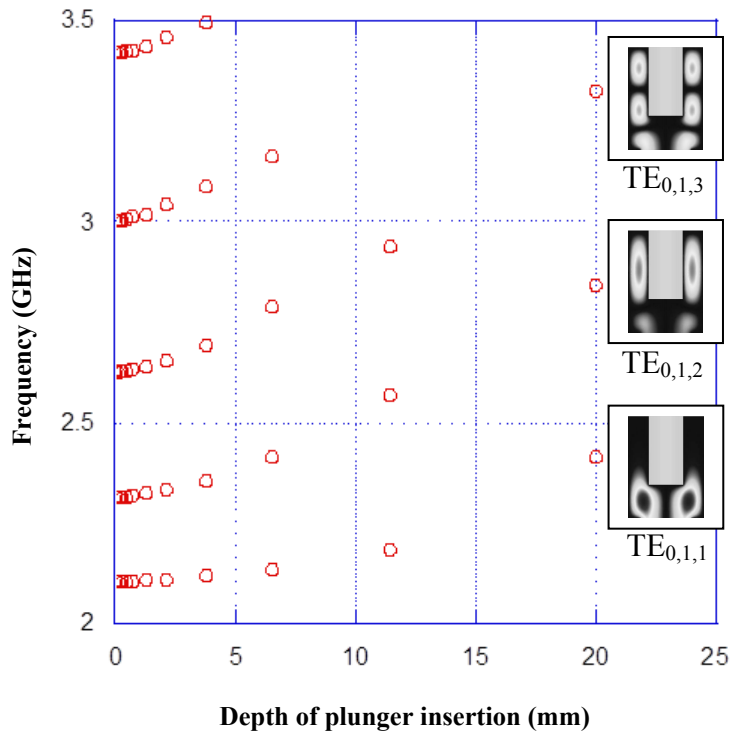


Figure 5: Illustration of the first electromagnetic resonances in the cavity and their frequency dependence to the depth of insertion of the metallic plunger.

To achieve our tuning requirements from 2.7 to 3.1GHz, we use the $TE_{0,1,3}$ resonance frequency mode, adjusting the frequency with the plunger. This technique prevents the need for using a deposition technique on the wafer with diamond, or employing a substrateless golden strip, which can easily be broken. The cavity delivers an homogeneous power, describing a ring of field over the diamond deposited wafer, which allows the interaction of many centers on a large scale, whereas the metal strip technique mimics an antenna and therefore the distribution of power is uneven from one end to another.

4. EXPERIMENTAL RESULTS

Figure 4 shows the lab-built confocal experimental setup we used for this experiment. The nanodiamonds containing NV centres are placed on a glass coverslip. The excitation laser (532 nm) is focused on the sample from below via a microscopy objective, which collects the light emitted by the NVs and redirects it back towards the detection path. The excitation light is cut off by mean of a notch filter to isolate the NV fluorescence signal. The detection path comprises a spectrometer and a Hanbury-Brown and Twiss (HBT) interferometer to respectively identify the NV centres and, amongst them, determine which ones are single emitters. The resonator carrying the microwave field to observe ODMR is placed above the sample and can be precisely moved along the three spatial axes via positioning screws. The ODMR spectrum is recorded via one of the photodetectors (APD-1 in Fig. 4) of the HBT interferometer.

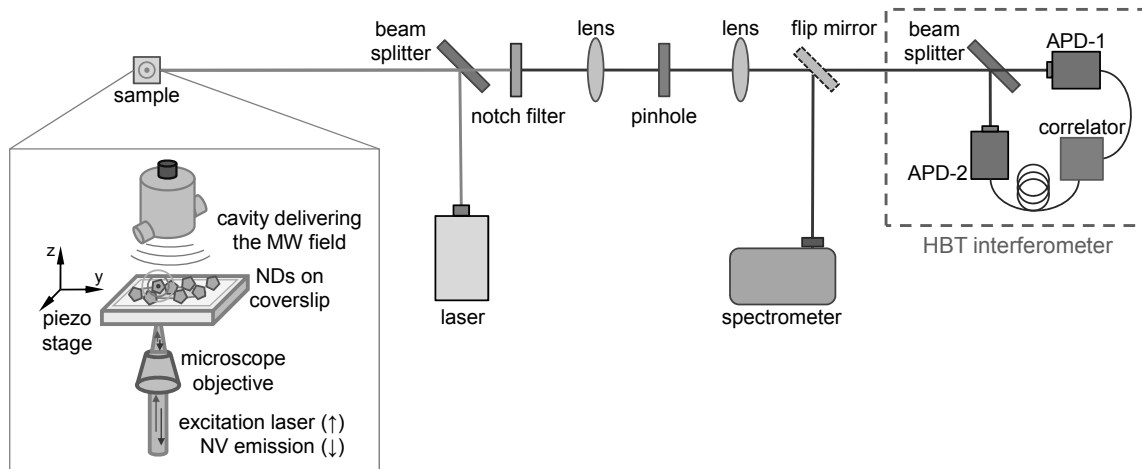


Figure 4: Schematic representation of the confocal experimental setup. The green laser (wavelength 532 nm) is focused on the sample. The sample consists of fluorescent diamond nanocrystals placed on a transparent coverslip mounted on a piezoelectric stage, while the microwave resonator to produce ODMR is placed above the nanodiamonds (inset). The fluorescence from the nanodiamonds is collected by the microscope objective, filtered via a notch filter to cut out the excitation light and directed to a pair of avalanche photodiodes (APDs) arranged in a Hanbury-Brown and Twiss (HBT) interferometer configuration. By switching a flappable mirror, the fluorescence is also collected by a spectrometer to identify the NV centres.

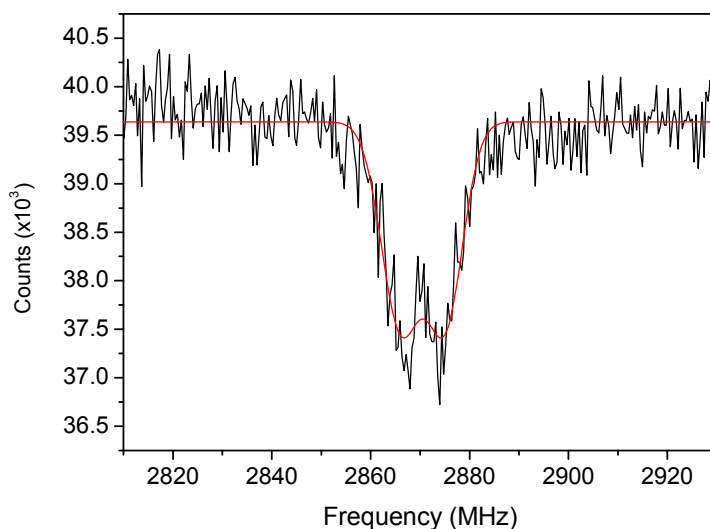


Figure 5: Continuous wave optically detected magnetic resonance (cwODMR) spectrum measured for a candidate NV centre. The relative change in fluorescence, while a microwave field is swept in the range 2810-2930 MHz, is recorded. The smooth curve superimposed on the data is a Lorentzian fit. The ground state sublevels $m_s = +1$ and $m_s = -1$ are slightly separated in energy due to strain in the nanodiamond crystal which breaks the NV trigonal symmetry. As a result, two distinguishable negative peaks can be observed in the spectrum.

Figure 5 shows the continuous wave ODMR spectrum measured on a single NV centre using the cavity. The relative change in fluorescence at around 2.87 GHz is clearly visible. Ideally, the two ground-state sublevels $m_s = +1$ and $m_s = -1$ are degenerate in energy and hence indistinguishable. The degeneracy is a consequence of the trigonal symmetry of the NV centre. Notice however how, in the figure, two peaks are almost distinguishable. This is typical of NV centres in nanodiamonds where the strong strain in the lattice causes a distortion in the trigonal symmetry of the centre and consequently lifts up the degeneracy.

5. CONCLUSIONS

The experiment conducted with the metallic microwave cavity tuned at 2.87 GHz has demonstrated it is possible to remotely drive the transition of the NV centre we are pumping to. This technique allows us to scan different centres where the microwave cavity field is radiating homogeneously over the nanodiamond deposited wafer. The contactless approach makes the ODMR much more flexible and requires less power to achieve the same result on a larger area defined by the cavity field propagation. Further work is going to study the application of this system on biological samples.

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