Optical determination and magnetic manipulation of a single nitrogen-vacancy color center in diamond nanocrystal

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Abstract
The controlled and coherent manipulation of individual quantum systems is fundamental for the development of quantum information processing. The nitrogen-vacancy (NV) color center in diamond is a promising system since its photoluminescence is perfectly stable at room temperature and its electron spin can be optically read out at the individual level. We review here the experiments currently realized in our laboratory concerning the use of a single NV color center as the single photon source and the coherent magnetic manipulation of the electron spin associated with a single NV color center. Furthermore, we demonstrate a nanoscopy experiment based on the saturation absorption effect, which allows to optically pin-point a single NV color center at sub-λ resolution. This offers the possibility to independently address two or multiple magnetically coupled single NV color centers, which is a necessary step towards the realization of a diamond-based quantum computer.

Keywords: nitrogen-vacancy, single photon source, single spin, superresolution

Classification numbers: 4.00, 4.03

1. Introduction

The nitrogen-vacancy (NV) color center in diamond has been identified as a system of choice for single photon generation [1] and individual electron spin manipulation [2] at room temperature. In particular, its electron spin has an exceptional long coherence time, which is advantageous over other systems envisaged for quantum computation operating at room temperature [3] and also for measuring magnetic field with high sensitivity and nanoscale resolution [4]. To this aim, a diamond nanocrystal (nanodiamond) containing a single NV color center is a key element, because it can be manipulated in space with high precision. On the other hand, a nanodiamond containing a NV color center can also be coupled to a photonic system such as a photonic crystal [5] resulting in enhancement of the single photon emission. However, for most of these applications it is important to know the orientation of the optical dipoles of the color center embedded in the nanodiamond, in order to efficiently couple the NV photoluminescence to the photonic structure. Such determination is directly related to spin orientation measurement since it has already been shown that the electron spin is orthogonal to the two independent optical dipoles involved in the photoluminescence. Spin orientation knowledge is useful to build up a vectorial ultra-sensitivity NV single-spin based magnetometer. Besides, it is also useful to be able to address optically and individually a single NV color center separated spatially from others at the nanometer scale. The study of two or multiple magnetically coupled NV electron spins will be the first step towards the realization of a diamond-based quantum computer operating at room temperature.

In this article, we first present the use of a single NV color center in nanodiamonds as an efficient single photon source.
Figure 1. (a) Experimental setup used to (i) address the individual color center, (ii) manipulate the single electron spin associated with the single NV color center and (iii) image NV color centers in diamond nanocrystals with sub-\(\lambda\) resolution. (b) Representative structure of the NV color center in the diamond matrix and schematic diagram of the NV ground-state electron spin energy levels.

at room temperature. Secondly, we demonstrate a simple technique based on the optically detected magnetic resonance to determine precisely the orientation of a single electron spin associated to a single NV color center embedded in an arbitrary nanodiamond. We finally demonstrate a microscopy experiment based on the saturation absorption effect, which allows us to optically pin-point single NV color centers at sub-\(\lambda\) resolution.

2. The experimental setup

The nanodiamond sample is prepared following a procedure described in [1], starting from type Ib synthetic diamond powder (de Beers, The Netherlands). The nanocrystals (about 90 nm mean diameter) are spin-coated on a dielectric mirror or on a glass substrate. The orientation of the nanodiamond crystallographic axis relative to the substrate surface is randomly distributed. Due to the \(C_{3v}\) symmetry of the NV centers in the diamond crystal, a single NV center is aligned along one of four possible orientations [6, 7] associated to the [111] axis of the crystal, which is un-determined due to the arbitrary orientation of the nanodiamond.

Optical excitation and detection of the NV color center photoluminescence is realized with a home-built setup, illustrated in figure 1(a). The NV color center is excited with a continuous laser (wavelength = 532 nm) and emits a broadband photoluminescence (about 100 nm—FWHM spectral width) centered at 670 nm. The excitation beam is tightly focused on the sample using an oil immersion high numerical aperture microscope objective (Obj). The focus point is raster scanned relative to the sample with nanometer resolution, using a 3D piezoelectric translation system. The photoluminescence of the excited NV center is collected by the same objective and spectrally filtered from the remaining pump light by a long-pass filter with wavelength cut-off at 580 nm. A standard confocal detection system (not shown) is also used to select the luminescence coming from a sample volume of about 1 \(\mu\)m\(^3\), ensuring the collection of light emitted by only one nanocrystal. The photoluminescence signal is finally detected by a silicon avalanche photodiode (APD) operating in the single-photon counting regime.

To apply the microwave field for electron spin resonance (ESR), the sample is placed on a circuit board with a strip line for microwave field input-output connection. A 20 \(\mu\)m-diameter copper wire placed over the sample is soldered to the strip lines. The wire is positioned within 20 \(\mu\)m of the optically addressed NV color center. The microwave power injected into the strip lines is fixed at 25 dBm for all measurements. The ESR measurement is then realized by sweeping the microwave frequency with a step of 0.3 MHz per channel, while detecting the corresponding luminescence photons. The external static magnetic field (\(B\)) applied to the NV center can be varied in magnitude and in orientation by moving and rotating a permanent magnet, which is mounted on three-axis translation and rotation stages, with respect to the sample.

In order to image the NV color centers with sub-\(\lambda\) resolution, a green excitation beam is passed through a phase mask (vortex phase plate), which enables a doughnut spot to be obtained at the focusing region of the microscope objective. The phase mask position is precisely controlled using a two-axis translation stage. The use of this phase mask necessitates high excitation power and it is used only for sub-\(\lambda\) imaging experiments.

3. Generation of a single photon from a single NV color center in a diamond nanocrystal

A single NV color center in diamond is a promising candidate among different kinds of solid-state single-photon sources due to the high efficiency and perfect photostability. Consisting of a substitutional nitrogen atom and a vacancy in an adjacent lattice site, the NV color center well absorbs an excitation light at 532 nm-wavelength and emits broadband...
photoluminescence with a maximum at around 670 nm and with a zero-phonon line at 637 nm. This color center plays the role of an atom or a molecule, which absorbs a single photon and emits only one photon at a time.

In order to realize such a single photon source, the sample is first scanned over a large area. A photoluminescence image is obtained as shown in figure 2(a), in which each bright spot represents the photoluminescence of NV color centers embedded in one nanodiamond. We then reposition the excitation beam and excite only one nanodiamond. To know if the nanodiamond contains only a single NV color center, the emitted photoluminescence is sent to a so-called Hanbury Brown and Twiss system, which separates a photoluminescence beam into two beams detected by two APDs (figure 2(b)). If the emission is single photon, no coincidence detection can be obtained because one photon cannot be split into two. Some of the nanodiamonds, which are numbered C3, C4 and C5 in figure 2(a), are therefore demonstrated to possess only a single NV color center, as indicated by the zero-coincidence detection by the two APDs shown in figure 2(c), at zero delay. A single photon source at room temperature is then obtained by exciting a well-determined nanodiamond. A triggered single-photon source was also obtained [1] by using the pulsed excitation of a single NV color center in a nanodiamond.

Note that for the ESR experiment, a single photon emission is always verified before all ESR measurements in order to magnetically manipulate the single electron spin associated with a single NV center.

4. Optically detected magnetic resonance of a single NV color center electron spin

In our sample, diamond nanocrystals mostly contain negatively charged NV color centers. The ground state (3A2) of the NV center is known to have an electron spin triplet structure with zero-field splitting of 2.87 GHz between the \( m_S = 0 \) and the degenerate \( m_S = \pm 1 \) states [6, 7]. The continuous laser excitation at 532 nm wavelength optically prepares the electron spin in the brighter \( m_S = 0 \) state (see figure 1(b)). When the microwave is applied to the NV center and its frequency is resonant with one of the spin transitions \( m_S = 0 \leftrightarrow m_S = \pm 1 \), the population is redistributed between the two levels, and the photoluminescence intensity decreases, as shown in figure 3(a). The determination of the spin resonance via the modulation of the photoluminescence intensity is therefore called optically detected magnetic resonance (ODMR).

We now apply this ODMR technique to precisely determine the orientation of a single NV color center embedded in an arbitrarily oriented nanodiamond, relative to the laboratory reference frame. Firstly, we apply the \( B \)-field along the Oz-axis (vertical), and we change \( B \)-magnitude by translating the permanent magnet along this Oz-axis (see figures 1(a) and 3(c)). By scanning the microwave frequencies, we obtained two resonance peaks for each magnet position (figure 3(b)), due to the Zeeman effect, with an increasing splitting upon the increase of the \( B \)-field magnitude (figure 3(c)). By comparing with the numerical simulation using the equation governing the spin Hamiltonian of the system, we infer the \( \theta \)-angle (57°) between the \( B \)-field and the NV spin orientation. With only the \( \theta \)-angle, the NV spin can be in any direction belonging to a cone with \( Oz \) as its symmetrical axis.

We then keep the distance between the magnet and the NV center constant in order to keep constant the \( B \)-magnitude applied to the NV spin. We rotate the magnet around the Oz-axis, by an angle \( \phi \), with the condition that the magnet North-South axis is always pointing toward the NV center (figure 3(d)). This results in a change of angle between the \( B \)-field and spin axis. According to the Zeeman effect, the resonance frequencies, corresponding to \( m_S = 0 \leftrightarrow m_S = -1 \) and \( m_S = 0 \leftrightarrow m_S = +1 \) transitions, vary as a function of \( \phi \). When the angle between the \( B \)-field and the spin axis angle is minimal (corresponding to \( \phi = 105^\circ \), the azimuthal angle), the splitting between the two resonance peaks is maximal.

By realizing these two measurements, we determined accurately in the laboratory frame the orientation of a single electron spin associated to a single NV color center in an arbitrary diamond nanocrystal [7].

5. Sub-diffraction-limited optical imaging of diamond nanocrystals

For NV color centers in diamond, the photoluminescence emission is saturated at rather low excitation power (about 1 mW, corresponding to a saturation intensity of about
Figure 3. Coherent manipulation of the single electron spin associated with a single NV color center in a diamond nanocrystal. The electron spin resonance spectrum obtained optically with $B = 0$ mT (a) and with $B \neq 0$ mT (b). (c, d) Determination of the orientation of a single NV spin by the ODMR method: (c) first measurement: the $\theta$-angle is obtained by changing the magnetic field magnitude, which is controlled by vertically translating the magnet along the $Oz$-axis; (d) second measurement: the $\phi$-angle is obtained by changing the magnetic field orientation. The magnet is rotated horizontally around the $Oz$-axis.

Figure 4. Comparison of luminescence images obtained by a commonly used confocal microscope and a negative Stimulated Emission Depletion (STED) microscope. (a, d) Focusing spots of the excitation beam. (b, e) Large area photoluminescence images. (c, f) Zoom in photoluminescence image, obtained at 13 mW excitation power. The FWHM of the dark spot is about 40 nm, which is much smaller than the diffraction limit. Two NV color centers, separated by 250 nm, are therefore clearly identified by a STED technique while they are un-separated with a confocal microscope.

$10^5 \text{ W cm}^{-2}$, i.e. the emission rate does not increase for excitation power above 1 mW. By using a doughnut-shaped excitation beam, the saturation is only reached in the doughnut region while the doughnut center remains dark, i.e. there is no excitation and no emission at the doughnut center. The size of the dark spot can become nanometric if the excitation power is very high compared to the excitation saturation level. By scanning the diamond nanocrystal sample using the
doughnut beam, we could therefore precisely determine the position of each NV color center, which is represented in the photoluminescence image by a dark spot. Figure 4 shows the photoluminescence images of NV color centers obtained with a Gaussian excitation beam (standard confocal microscope) and with a doughnut excitation beam (STED-like microscope [8]). It is difficult to distinguish the position of NV color centers using a commonly used confocal technique. In contrast, when using the doughnut-shaped excitation beam, each NV color center is represented by a dark spot and distinguished from the others. Moreover, with an excitation power of 13 mW, the dark spot becomes effectively nanometric in size, and two NV color centers separated by only about 250 nm are clearly identified (figure 4(f)). This imaging technique is effectively very useful to determine the position of NV color centers, with precision well below the diffraction limit of regular confocal microscopy.

In order to achieve an even higher resolution, it is necessary to improve the quality of the doughnut beam, i.e. optimize the phase mask for the excitation wavelength. Besides, to address optically a single color center without perturbing the neighbors and to directly obtain the photoluminescence from the nanospot, the realization of a so-called positive STED will be necessary, by considering for example two alternative excitations, one with a Gaussian form and another with doughnut form [8]. The success of this imaging technique will open the way to single spin optical addressing in the realization of diamond-based quantum computer functioning at room temperature.

6. Conclusion

Our work focused on the study of fundamental physical properties of NV color centers in diamond nanocrystals and applications based on the optical and magnetic properties. The NV color center is demonstrated to be an excellent single photon source, reliable and stable at room temperature, which can be used for many applications, such as quantum experiments or quantum key distribution. The single electron spin associated with a single NV color center is coherently and optically manipulated and it now becomes an excellent candidate for quantum information processing, in particular for building up a new ultrasensitive magnetometer. A super-resolution imaging technique is also demonstrated, based on the use of a doughnut-shaped excitation beam at high excitation power. This enables optical addressing of individual spins to be carried out in a future experiment of coupled multiple electron spins, which paves the way to a diamond-based quantum computer.

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