

Excited-state spectroscopy of single NV defects in diamond using optically detected magnetic resonance

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Abstract. Using pulsed optically detected magnetic resonance techniques, we directly probe electron-spin resonance transitions in the excited-state of single nitrogen-vacancy (NV) color centers in diamond. Unambiguous assignment of excited state fine structure is made, based on changes of NV defect photoluminescence lifetime. This study provides significant insight into the structure of the emitting ³E excited state, which is invaluable for the development of diamond-based quantum information processing.

Over the last decade, the negatively charged nitrogen-vacancy (NV) color center in diamond has attracted a lot of interest because it can be optically addressed as single quantum system [1] and exhibits several important properties for quantum information science applications. Firstly, its perfect photostability at room temperature enables a practical NV-based single photon source [2, 3] to be realized for quantum cryptography applications [4, 5]. Secondly, NV color centers have a paramagnetic ground state whose spin can be optically polarized, read-out and exhibits long coherence time even at room temperature [6, 7]. Coherent manipulation of electron and

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nuclear spins of single NV color centers has been used to realize solid-state quantum physics experiments, ranging from coherent coupling of a single NV color center to other single spins in the diamond crystalline matrix [7]–[9], to the implementation of a quantum register [10] and conditional two-qubit CNOT gates [11], and very recently the generation of Bell and GHZ states with long coherence times [12].

Despite these results, which make the NV color center a competitive candidate for solid-state quantum information processing, the excited-state structure of the defect is not yet fully understood [13, 14]. This knowledge is, however, crucial for single-spin high-speed coherent optical manipulation through Λ -based transitions [15]–[17] as well as for future implementation of quantum information protocols like quantum repeaters [18, 19] that can be used as a building block for a quantum network [20].

Resonant optical excitation of single NV color centers at low temperature [21] and continuous wave (cw) electron-spin resonance (ESR) experiments [22] have recently provided new insights into the structure of the excited-state, showing that its fine structure is strongly affected by local strain in the diamond matrix [21, 22]. Recent ensemble experiments have also studied the behavior of an infrared emission line that gives a better understanding of the metastable state responsible for spin polarization [23].

Here, we develop a new approach to probe the excited-state fine structure of single NV color centers. Using pulsed optically detected magnetic resonance (ODMR) techniques, we directly probe ESR transitions of the excited-state. Unambiguous assignment of excited state fine structure is made, based on changes of NV defect photoluminescence lifetime.

The NV defect center in diamond consists of a substitutional nitrogen atom (N) associated with a vacancy (V) in an adjacent lattice site of the diamond crystalline matrix (figure 1(A)). For the negatively charged NV color center, which is addressed in this study, the ground state is a spin triplet state 3A , originating from six unpaired electron spins. Owing to the C_{3v} symmetry of the NV center, ground state spin sublevels $m_s = \pm 1$ are degenerate and the zero-field splitting from $m_s = 0$ is $D_{gs} = 2.87$ GHz (figure 1(B)). The excited state 3E is also a spin triplet, associated with a broadband photoluminescence emission with zero phonon line at 1.945 eV. The order of other energy levels is still under debate, but it is now well established that at least one metastable state 1A lies between the ground and excited triplet state [13, 23]. This metastable state plays a crucial role in spin dynamics of the NV color center. Indeed, whereas the optical transitions $^3A \rightarrow ^3E$ are spin conserving, non-radiative inter-system crossing transitions to the metastable state $^3E \rightarrow ^1A$ are strongly spin selective as the shelving rate from the $m_s = 0$ sublevel is much smaller than those from $m_s = \pm 1$. Conversely, the metastable state decays preferentially towards the ground state $m_s = 0$ sublevel, leading to a strong spin polarization into $m_s = 0$ after a few optical excitation–emission cycles [24].

Another consequence of this spin-selective process is that the photoluminescence intensity is higher when the $m_s = 0$ state is populated, allowing optical detection of spin-rotation of a single NV center at room temperature by ODMR [25, 26]. Indeed, if a single NV center, initially prepared in the $m_s = 0$ state through optical pumping, is driven to the $m_s = \pm 1$ spin sublevels by applying a resonant microwave frequency, a decrease in photoluminescence signal is observed. This technique is now routinely used for single-spin readout in solid-state quantum optics experiments using single spins in diamond as quantum bits. Until now only the ground state spin sublevels have been detected using ODMR. However, as optical transitions are spin conserving, spin rotations in the excited states should be also detected, allowing the structure of spin sublevels to be probed.

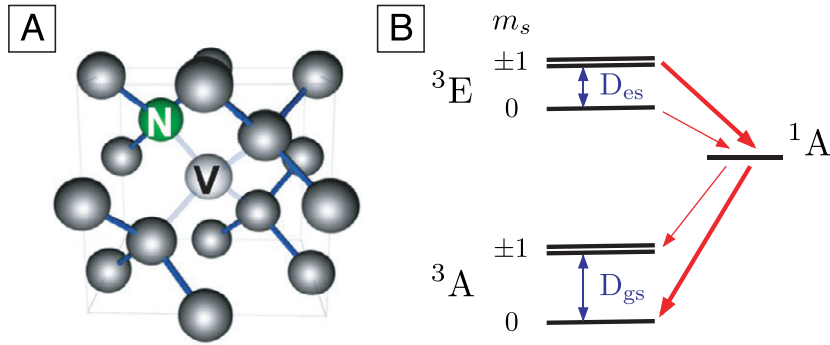


Figure 1. (A) Atomic structure of the NV center. The single substitutional nitrogen atom (N) is accompanied by a vacancy (V) at a nearest neighbor lattice position. (B) Simplified energy-level diagram of the NV center. D_{gs} and D_{es} correspond, respectively, to the zero-field splitting between $m_s = 0$ and $m_s = \pm 1$ in the triplet ground state 3A and in the triplet excited state 3E . Spin selective shelving into a metastable singlet state 1A (red arrows) leads to spin-polarization of the center into the ground state $m_s = 0$ sublevel through optical pumping.

We first investigate single NV color centers artificially created in a ultra-pure type IIa diamond sample (Element6), by implanting 7 MeV isotopically pure ^{15}N atoms and by annealing the sample for 2 h in vacuum at 800 °C [27]. NV centers are then optically addressed at room temperature using a standard confocal microscope coupled to a Hanbury-Brown and Twiss setup used to measure the photoluminescence second-order correlation function $g^{(2)}(\tau)$ and verify that an individual NV center is addressed.

ESR spectroscopy of single NV centers is realized by applying microwaves, using a copper microwire (20 μm diameter) close to the NV center, and by monitoring the photoluminescence intensity. When the microwave frequency is resonant with the transition between $m_s = 0$ and $m_s = \pm 1$ sublevels, spin rotation is evidenced as a dip of the photoluminescence signal as explained above. Figure 2(A) (upper trace) describes a typical ODMR signal obtained by sweeping the microwave frequency without any applied magnetic field. The well-known ground state transition between $m_s = 0$ and $m_s = \pm 1$ sublevels is detected at 2.87 GHz, and an additional broad line around 1.4 GHz is observed. By applying a magnetic field to the sample, the degeneracy of $m_s = \pm 1$ is lifted by the Zeeman effect, leading to the appearance of two lines in corresponding resonances of the ODMR spectrum (see figure 2(A) bottom trace). As a hypothesis, we attribute the broad resonance to spin sublevels of an excited state of the NV color center. In the following, we will demonstrate that this excited state actually corresponds to the emitting excited state 3E .

We first study in more detail the ESR frequency positions as a function of the magnitude of a magnetic field (B) applied along the NV symmetry axis which corresponds to a [111] crystal axis. The results of this experiment are depicted in figure 2(B). Neglecting electron–nuclear spin coupling, the excited-state spin Hamiltonian of the NV defect can be written as

$$H = D_{es}(S_z^2 - \frac{2}{3}) + E_{es}(S_x^2 - S_y^2) + g_{es}\mu\vec{B} \cdot \vec{S}, \quad (1)$$

where D_{es} is the excited-state zero-field splitting, $S = 1$, E_{es} is the excited-state strain-induced splitting coefficient, g_{es} the excited state g -factor and μ the Bohr magneton. By considering

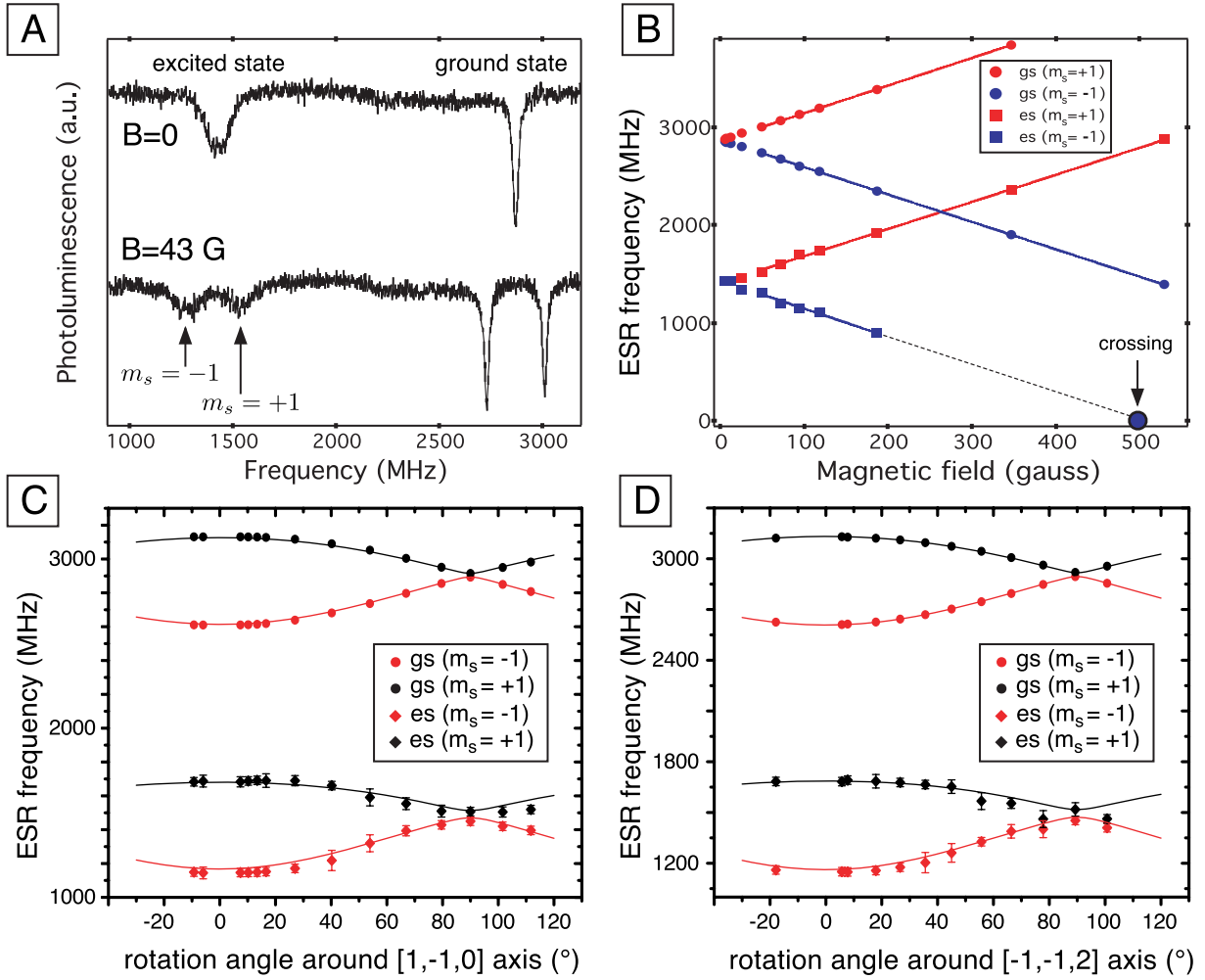


Figure 2. (A) ODMR spectra of a single NV color center at zero magnetic field (upper trace) and with a magnetic field of amplitude $B = 43$ G applied along the NV symmetry axis which corresponds to a $[111]$ crystal axis (bottom trace). ESRs are evidenced both in the ground state (gs) and in the excited state (es). (B) ESR frequencies as a function of the amplitude of the applied magnetic field along a $[111]$ crystal axis. Solid lines correspond to a fit of the experimental results using equation (2), leading to $D_{\text{es}} = 1423 \pm 10$ MHz and $g_{\text{es}} = 2.01 \pm 0.08$. A level anti-crossing between the excited state $m_s = 0$ and $m_s = -1$ sublevels is expected for a magnetic field around 500 G. (C) and (D) ESR frequencies measured by rotating a magnetic field of amplitude $B = 92$ G around a $[1, -1, 0]$ crystal axis (C) and around a $[-1, -1, 2]$ crystal axis (D). The solid lines correspond to theoretical expectations using the Hamiltonian described by equation (1) with the measured values of D_{es} and g_{es} , and neglecting the strain-induced splitting E_{es} .

magnetic field magnitudes such that the strain-induced fine structure splitting is negligible compared with Zeeman splitting ($|E_{\text{es}}(S_x^2 - S_y^2)| \ll |g_{\text{es}}\mu\vec{B} \cdot \vec{S}|$), the resonant frequencies

ω_{\pm} associated to eigenstates $m_s = \pm 1$ are given by

$$\omega_{\pm} = D_{\text{es}} \pm g_{\text{es}}\mu B. \quad (2)$$

For the NV center considered in this study, it was not possible to observe any strain-induced splitting in the excited state, the ODMR dip being very broad with a FWHM on the order of 100 MHz (see figure 2(A)). As a result, it is reasonable to consider that the strain-induced splitting coefficient is such that $2E_{\text{es}} \ll 100$ MHz.⁴ Following this consideration, the experimental results depicted in figure 2(B) can be fitted using equation (2) for magnetic field magnitudes bigger than 50 G, corresponding to a Zeeman splitting on the order of 130 MHz ($\gg E_{\text{es}}$). The results of the fit lead to $D_{\text{es}} = 1423 \pm 10$ MHz and an isotropic g -factor $g_{\text{es}} = 2.01 \pm 0.08$ which is similar to the ground state g -factor. This isotropy indicates that the orbital angular momentum does not play a significant role in the excited state. Finally, the positions of ESR frequencies were measured by rotating a magnetic field of magnitude $B = 92$ G around a $[1, -1, 0]$ crystal axis (figure 2(C)) and around a $[-1, -1, 2]$ crystal axis (figure 2(D)). The experimental results provide strong evidence that the ground and excited states exhibit the same orientations.

It is interesting to note that a level anti-crossing between the $m_s = 0$ and $m_s = -1$ sublevels of the excited state is expected for a magnetic field amplitude on the order of 500 G (see figure 2(B)). This explains why a decrease of the NV color center photoluminescence has been observed in ensemble experiments at such magnetic fields [23, 28]. Indeed, when a level anti-crossing occurs in the excited state, the electron spin polarization of the center is significantly reduced. This effect is identical to the well-known photoluminescence dip occurring at $B = 1028$ G, when the $m_s = 0$ and $m_s = -1$ sublevels of the ground state are crossing.

The cw experiments reported above prove that the ESR signal corresponds to an excited state, but do not provide ultimate proof that this state is responsible for fluorescence emission. For example, other dark states involved in the spin polarization pathway can influence the spin polarization of NV defects [23]. We now demonstrate that the excited state observed in ODMR spectra actually corresponds to the emitting excited state ^3E . Following a method introduced in [29], the experiment is based on observing a modification of photoluminescence decay by manipulating the excited state spin sublevels with resonant microwave pulses.

For that purpose, a large microwave driving field is required in order to reach a significant change in the population of the excited-state spin sublevels within the radiative lifetime. In order to more easily meet that requirement, we used single NV color centers in diamond nanocrystals, for which the photoluminescence decay is known to be much longer than in bulk diamond samples [30]. In addition, the nanodiamonds were spin-coated on a microscope cover glass on which gold strip-line microwave wires had been deposited using shadow mask photolithography and metal electrodeposition in order to reach high ESR Rabi frequencies. Typical dimensions of the wires were $10\ \mu\text{m}$ width and $2\ \mu\text{m}$ thick. Figure 3(A) shows a photoluminescence map of the sample, the yellow cursor indicating the individual NV center studied in the following. For this NV center, we measured a Rabi nutation between ground state spin levels at a frequency of 200 MHz, corresponding to a π pulse of 2.5 ns (figure 3(B)).

As depicted in figure 4(A), the ODMR spectrum shows a huge energy splitting between ESR lines, even at small magnetic field magnitude ($B = 20$ G). Such splitting, which cannot be explained by the Zeeman effect, results from strain-induced splitting which is known to be

⁴ Note that surprisingly, for most of the single NV centers ($\approx 80\%$) studied in our implanted sample it was not possible to observe a strain-induced splitting in the excited-state.

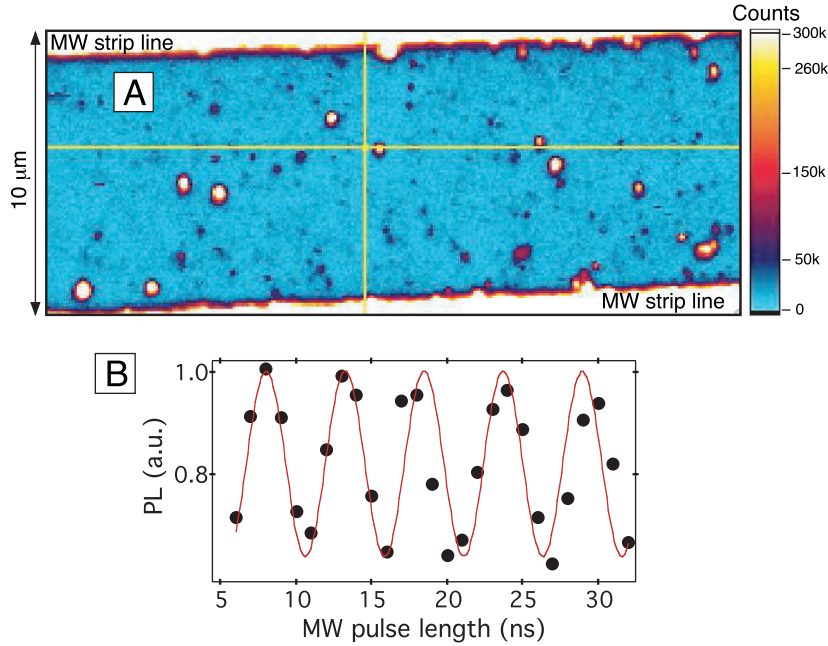


Figure 3. (A) Confocal fluorescence image of the sample showing strip-line microwave wires and NV centers in diamond nanocrystals. Fluorescence intensity is encoded in color scale and the yellow cursor indicates the NV center of interest, which was verified as a single emitter using photon correlation measurements (data not shown). (B) Rabi nutation between ground state spin levels of this single NV center, indicating a 2π rotation within 5 ns.

much stronger in diamond nanocrystals than in bulk samples [31]. However, note that strain has almost no effect on the D values (D_{gs} and D_{es}), whereas it does cause the E values (E_{gs} and E_{es} in the Hamiltonian described by equation (1)) to become nonzero. In the following, we use resonant microwaves with the ground (MW_{gs}) and excited state (MW_{es}) $m_s = 0 \rightarrow m_s = -1$ transitions, at 2844 and 1000 MHz, respectively (see figure 4(A)).

As a first step, the excited state lifetime associated with each spin sublevel was measured. The NV center was first polarized into the ground state $m_s = 0$ sublevel using an optical pulse of duration $3 \mu\text{s}$ at the wavelength $\lambda = 532 \text{ nm}$. After a time delay of $1 \mu\text{s}$, which ensures that the NV center has relaxed to the ground state, an optical pulse (40 ps , $\lambda = 532 \text{ nm}$) much shorter than the radiative lifetime was used to excite the NV center. As the optical transition is spin-conserving, such a sequence allows to build up the photoluminescence decay of the excited-state $m_s = 0$ sublevel using a standard start–stop technique for lifetime measurements, the start being a part of the pulsed excitation and the stop being the single-photon counter signal. For measuring the decay of the $m_s = -1$ sublevel, an additional microwave π pulse resonant with ground state spin transition (MW_{gs}) was introduced before the ps optical pulse.

The results of such measurements are depicted in figure 4(B). Fluorescence decays follow single exponential decay associated with a lifetime $\tau_0 = 23 \text{ ns}$ for the $m_s = 0$ sublevel and $\tau_{-1} = 12.7 \text{ ns}$ for $m_s = -1$. As expected, these values are much bigger than the ones measured in bulk samples [29]. As the photoluminescence is always smaller from $m_s = \pm 1$ sublevels compared to $m_s = 0$, the measured lifetime difference results from spin-selective non-radiative

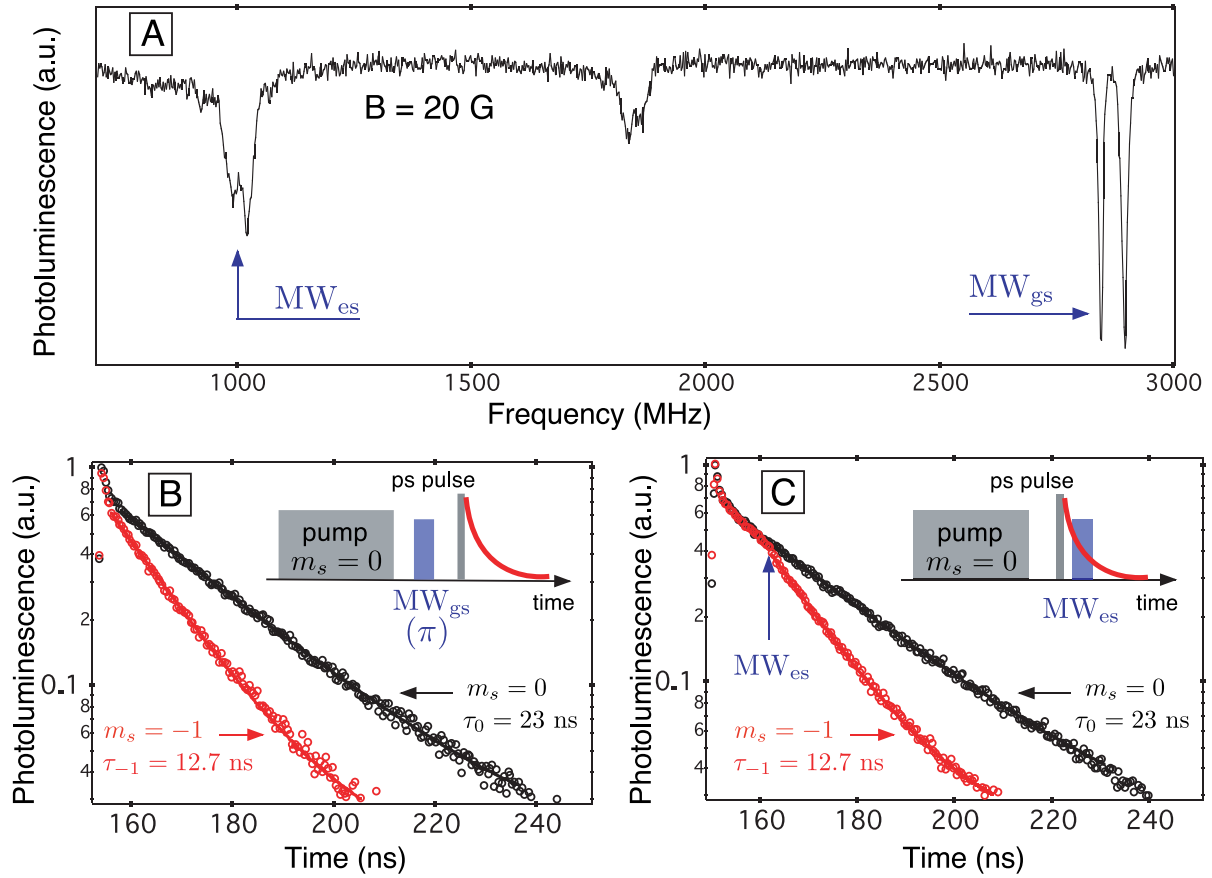


Figure 4. (A) ODMR spectra of the studied single NV color center in a diamond nanocrystal recorded with a 20 G magnetic field applied. MW_{gs} and MW_{es} , respectively, correspond to the resonant frequency of the $m_s = 0 \rightarrow m_s = -1$ transition in the ground state and in the excited state. Note that careful examination of the excited state resonances shows multiple dips which might be related to excited-state hyperfine structure [22]. (B) and (C) Spin-selective photoluminescence decay curves, monitored by applying a spin selective resonant microwave pulse (B) in the ground state (frequency MW_{gs}) and (C) on the excited state (frequency MW_{es}). The insets describe the time sequence of the experiments (see main text) and solid lines are single exponential fit curves, leading to a radiative lifetime $\tau_0 = 23$ ns for the $m_s = 0$ sublevel and $\tau_{-1} = 12.7$ ns for $m_s = -1$.

inter-system crossing transition to the metastable state, and not from a modification of the transition strength. It is interesting to notice that the measured lifetime ratio, $\tau_{-1}/\tau_0 \approx 0.55$, is in good agreement with recent theoretical predictions [13] and previously reported measurements in bulk samples [29].

In order to check if the resonance lines observed in ODMR spectra actually correspond to spin transitions in the emitting 3E excited state, the same experiment is performed by applying a microwave pulse resonant with the excited state spin transition $m_s = 0 \rightarrow m_s = -1$ (MW_{es}), just after the picosecond optical pulse. The results, depicted in figure 4(C), indicate a drastic change

of the photoluminescence decay when the microwave pulse is applied. Before the microwave excitation, the photoluminescence decay follows the single exponential decay associated with the excited-state $m_s = 0$ sublevel ($\tau_0 = 23$ ns). The microwave excitation then suddenly rotates the spin in the excited state leading to the exponential decay associated with the excited-state $m_s = -1$ ($\tau_{-1} = 12.7$ ns). These results unambiguously evidence that the new ESR lines observed in ODMR spectra are related to fine structure of the emitting excited state 3E .

It was not possible to detect Rabi nutations on the excited state by varying the resonant microwave pulse duration. This appears as a difficult issue because of many factors. Among them are the large width of the excited-state ESR resonance, a strong hyperfine coupling owing to high spin density of the excited-state wavefunction at the nitrogen nucleus [32] and the short radiative lifetime of NV color centers.

Finally, we would like to briefly discuss our observations in the context of previously reported models of the excited-state structure. Resonant optical excitation of single NV color centers at low temperature has recently indicated that the excited-state is actually an orbital doublet, split into two orbital singlet branches by local strain [21]. Then, we would expect to observe four excited-state resonances when a magnetic field is applied to the NV center, while we only ever observed two lines (figures 2(A) and 4(A)). We tentatively identify the observed excited-state features as the upper branch because this branch shows high difference in inter-system crossing rate to the metastable state [13, 29], as observed in experiments (see figure 4(B) and (C)). However, a previous model of the excited-state structure has predicted non-vanishing E_{es} in the upper branch [21]. The observations in bulk diamond reported in this paper ($E_{es} \approx 0$) bring then into question the completeness of currently available models for NV center excited-state structure.

Summarizing, we have performed the excited-state spectroscopy of single NV color centers in diamond using cw and pulsed ESR techniques. This work provides significant insight into the structure of the emitting 3E excited state, which might be useful for diamond-based quantum information processing using Λ -based transitions for high-speed coherent optical manipulation of single spins [17] as well as for entanglement protocols used in quantum repeaters [18, 33].

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