Two-quantum optically detected resonances in NV centers in diamond in zero magnetic field

Alexander K. Dmitriev, Anton K. Vershovskii

Ioffe Institute, 26 Politekhnicheskaya, St. Petersburg 194021 Russia
E-mail: alexdmk777@gmail.com

Introduction

The methods for controlling spin states of negatively charged nitrogen-vacancy centers using a combination of microwave (MW) or radiofrequency (RF) excitation field [1,2] for electron spin transitions and RF excitation field for nuclear spin transitions are most effective in strong magnetic fields where level anti-crossing (LAC) occurs [3]. However, LAC in zero field can also be used to control spin states, as well as to excite narrow resonances for metrological application. Here we present magnetically independent resonances arising in the ODMR spectra of NV centers in bulk diamond under two-frequency (MW+RF) resonant excitation in zero magnetic field, and discuss their specificity.

Energy structure of the ground-state of NV center

The level structure of $^3A_2$ ground state in external magnetic field $\vec{B}$ is defined by the Hamiltonian [4]:

$$H = D(S_z^2 - \frac{1}{3}\vec{S}^2) + E(S_x^2 - S_y^2) + g_s\mu_B\vec{B}\cdot\vec{S} + A_{\|}S_zI_z + A_{\perp}(S_xI_x + S_yI_y) + PI_z^2 - g_I\mu_N\vec{B}\cdot\vec{I}, \qquad (1)$$

where $\mu_B = h\cdot 13.996\cdot 10^9$ Hz/T is the Bohr magneton, $\vec{I}$ is the ^{14}N nuclear (I = 1), $\vec{S}$ is the electron spin of NV center (S = 1), $\mu_N = h\cdot 7.622\cdot 10^6$ Hz/T is the nuclear magneton, D = 2.87 GHz and E are axial and transverse zero-field splitting (ZFS) parameters, $g_s = 2.003$ and $g_I = 0.403$ are electron and nuclear g-factors, $A_{\|} = -2.16$ MHz and $A_{\perp} = -2.7$ MHz are axial and transverse hyperfine splitting parameters, P = 4.95 MHz is the quadrupole splitting parameter. Denote eigenstates of the ground state $|m_S, m_I\rangle$; for the nitrogen isotope ^{14}N both electronic and nuclear spin projections take values $m_S, m_I = 0, \pm1$.

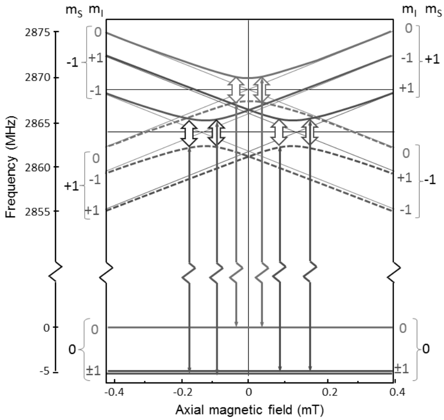

Figure 1. NV center ground-state splitting frequencies' dependence on axial local magnetic field, calculated for a diamond crystal with transverse ZFS parameter E = 1.8 MHz. Single arrows represent the MW drive field, double arrows represent the RF drive field

The energy structure of NV center in zero and ultra-weak fields is more complex than in strong ones (Fig.1); it contains both level crossings and anti-crossings, partially masked by the inhomogeneity of the crystal's internal fields. Therefore, pure energy states $|m_S, m_I\rangle$ at $B \approx 0$ mix in superpositions.

Experiment and discussion

The experimental setup was described in [5]: a synthetic diamond of SDB1085 60/70 grade (manufactured by Element Six) with dimensions 0.1 × 0.3 × 0.3 mm was subjected to electron irradiation ($5{\cdot}10^{18}$ cm^{-2}) and subsequent annealing in Ar at 800°C over 2 hours. The crystal was used at room temperature; it was attached by optically transparent glue to the end of an optical fiber.

We excited two-frequency ODMR in $B = (0 \div 1)$ mT using MW drive field f_{MW} in combination with additional RF field f_{RF}. This way we have recorded two symmetrical hollows in ODMR spectrum (Fig.2, 3), arising under conditions

$$f_{MW} \pm ½ f_{RF} = D, \tag{2}$$

$$\begin{gathered} \nu_0 - \Delta < 2|f_{MW} - D| < \nu_0 + \Delta, or \\ \nu_0 - \Delta < f_{RF} < \nu_0 + \Delta, \end{gathered} \tag{3}$$

where $\nu_0 = (4.34 \pm 0.02)$ MHz is the center of the resonance envelope in RF scale, and $\Delta = (2.14 \pm 0.04)$ MHz is the half-width of the envelope. We have observed similar resonances previously, while applying low-frequency amplitude modulation to the MW field [6]; this time we applied the same modulation to the RF field, which caused at least a two-fold increase in the contrast of the resonances, and changed their shape.

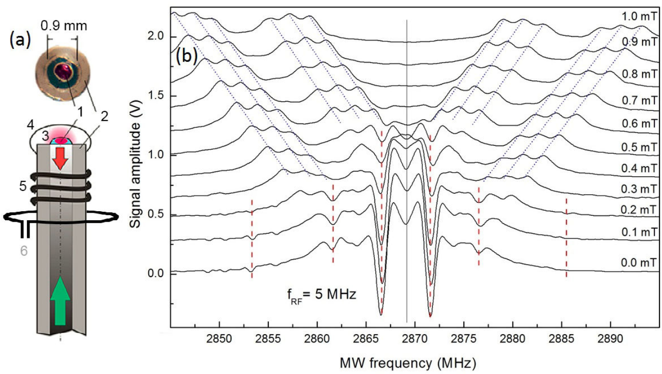

Figure 2. (a) View of the fiber end, and a schematic diagram of magnetometer sensor: 1 - diamond crystal, 2 - optical fiber, 3 - transparent glue, 4 - reflective coating, 5 - MW antenna, 6 – RF antenna. (b) ODMR spectra recorded at external field $B = (0 \div 1)$ mT along (1,1,1) direction with additional RF excitation at $f_{RF} = 5$ MHz

At the optimal (i.e. providing the maximal resonance steepness) values of MW and RF amplitudes, the linewidth (HWHM) of the resonances was found to be about 1.1 MHz. The amplitude of the peaks is maximal at zero magnetic field, and it decreases quickly as the field induction decreases (Fig. 2). Moreover, the frequencies of both resonances proved to be insensitive to B. On the other hand, the fact that according to (2) the combination of resonant MW and RF frequencies depends only on D makes these peaks very attractive for the task of frequency stabilization.

Conditions of resonance observation are typical for a two-quantum resonance, represented by vertical arrows on Fig. 1 (single arrows represent the MW drive field, double arrows represent the RF drive field); a variety of similar multi-frequency resonances arising due to excited state LAC in a strong (51 mT) field have been studied in [3]. However, the resonances discovered in zero field in our work show some peculiarities: they appear as dips in the "normal" one-quantum ODMR signal. Therefore, their nature must be similar to the "dark" resonances due to the coherent population trapping effect (CPT) in Λ-schemes [7].

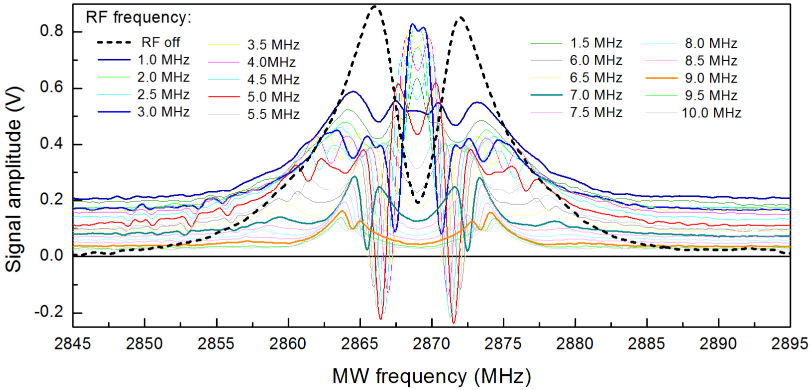

Figure 3. ODMR spectra recorded at zero external field at different radiofrequencies f_{RF}*; lowest curve is normal ODMR signal, two symmetrical hollows are two-quantum resonances arising when conditions (2), (3) are fulfilled*

Conclusion

We report the detection of high-contrast magnetically independent two-quantum resonances in zero-field ODMR spectra of NV center in diamond, induced by applying an additional modulated RF field. These resonances can only be driven at $|B| < 0.5$ mT (in the case of our diamond sample), and therefore we can assert that they are due to the zero-field level anti-crossing. We attribute them to certain transitions in NV center's zero-field structure.

References

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