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# Ultra-narrow low-field nuclear spin resonance in NV centers in bulk diamond crystal

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## Introduction

The application of methods based on optically detected magnetic resonance (ODMR) to negatively charged nitrogen vacancy ($NV^-$, or just NV) color centers in diamond crystals has brought forth relatively simple new methods of controlling nuclear spins. These spins are considered to be excellent candidates for solid-state quantum information processing because of their very long coherence time [1,2]. Typically, level anticrossing (LAC) [1,2] or combined microwave (MW) and radiofrequency (RF) excitation [3] are used to address chosen spin state, and the width of observed resonances exceeds hundreds of kilohertz. To reduce the resonance width, special techniques, such as single spin excitation, should be used. Here, however, we report on ultra-narrow (~7 kHz HWHM) resonances, which can be excited and optically detected in ultra-weak (≥1 G) magnetic fields using a single RF field ~4.95 MHz.

## Ground-state nuclear spin resonance

Level structure of $^3A_2$ state is defined by Hamiltonian [4,5]

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

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.

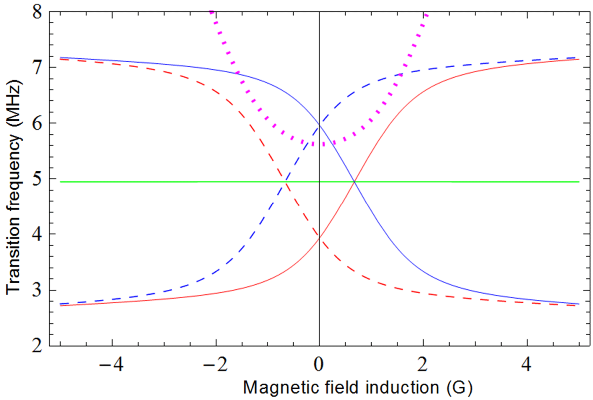


*Figure 1. Calculated frequencies of nuclear transitions in diamond crystal with E = 1.8 MHz. Red lines denote |1,1>↔|1,0> and |1,0>↔|1,−1> transitions (first digit is electronic, second – nuclear spin projection), green represents |0,0>↔|0,±1>, blue represents |−1,1>↔|−1,0> and |−1,0>↔|−1,−1>. Dotted magenta line represents a spin-flip electron-nuclear transition |1,−1>↔|−1,1>.*

Frequencies of nuclear transitions, calculated from (1) for diamond crystal with $E$ = 1.8 MHz, are shown in Fig. 1. All of these transitions, save for the pure nuclear |0,0>↔|0,±1> transitions, are strongly dependent on the magnetic field, and therefore they should be broadened by an inhomogeneous magnetic field in the crystal.

It is not evident *a priori* that these resonances can observed using only RF excitation without MW: they can only appear in the ODMR spectrum under the condition that optical pumping creates a population difference between |0,0> and |0,±1> levels. Nevertheless, in our experiment we have recorded both nuclear |0,0>↔|0,±1> and spin-flip electron-nuclear |1,−1>↔|−1,1> transitions (Fig 2).

The experimental setup was described in [6]: 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.

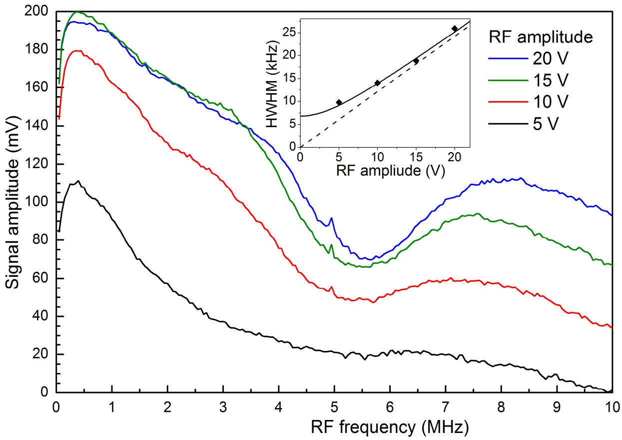


*Figure 2. RF ODMR spectra at zero field recorded at different RF amplitudes; inset - signal width dependency on RF amplitude at B = 10 G.*

RF ODMR spectra recorded at zero magnetic field (Fig.2) exhibit a) a wide hollow centered at ~5.6 MHz and therefore, according to Fig.1, with high certainty attributed to the spin-flip electron-nuclear transition, and b) a narrow peak centered at 4.95 MHz, corresponding to the |0,0>↔|0,±1> nuclear transition. The resonance linewidth dependency on RF amplitude was recorded at 10 G in order to maximize signal-to-noise (SNR) ratio (inset on Fig.2). Fitting shows that the linewidth (HWHM) extrapolated to zero RF amplitude is only 6.8 kHz, corresponding to transverse relaxation time $T_2^*$ = 23 μs. The ODMR signal cutoff frequency $f_l$ = 220 Hz is limited by the population relaxation constant: $T_1 \geq 1/(2\pi \cdot f_l)$ = 720 μs.

To check that observed resonances really correspond to the |0,0>↔|0,±1> nuclear transition, we recorded RF ODMR spectra in magnetic fields up to 100 G (Fig.3). The very low SNR ratio in Fig 3 is due to 1) noisy environment, and 2) the low RF amplitude chosen in order to avoid broadening the resonance line. Nevertheless, it is apparent that the value of the resonance splitting corresponds to the expected value of 2·307 Hz/G; the simultaneous shift in the resonance lines is yet to be explained.

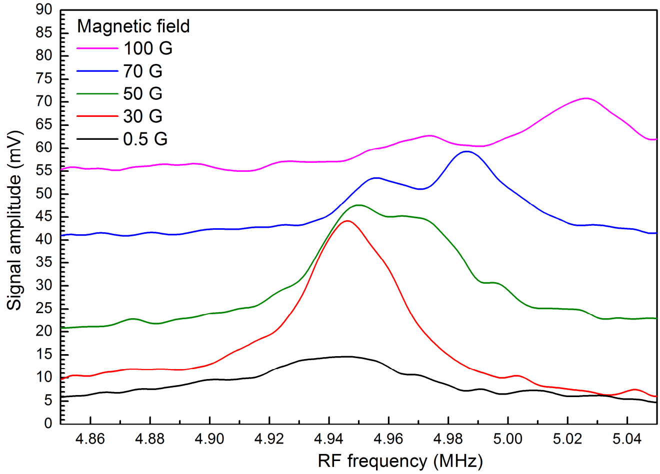


*Figure 3. RF ODMR spectra recorded at different magnetic field values.*

## Conclusion

Our experiment shows that ultra-narrow (<10 kHz) nuclear transition resonances can be observed in RF ODMR signal at room temperature in a low magnetic field without additional MW excitation; the signal-to-noise ratio of these resonances is subject to further improvement. This can be an important step to a simple and compact scheme of nuclear spin control for the task of quantum computation.

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