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High-frequency pulsed ENDOR spectroscopy of the NV^- centre in the commercial HPHT diamond

B.V. Yavkin *, G.V. Mamin, S.B. Orlinskii

Kazan Federal University, Institute of Physics, 420008 Kazan, 18 Kremlyovskaya St, Russian Federation

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ABSTRACT

This work reports direct 94 GHz ENDOR spectroscopy of the ^{14}N nuclei in the NV^- centre in single-crystal diamond. Roadmaps of ENDOR frequencies were measured and hyperfine/quadrupole interaction parameters were obtained, with $A_{X,Y} = -2.7$ MHz, $A_Z = -2.2$ MHz and $P = -4.8$ MHz. The sign and value of each parameter was calculated using spin Hamiltonian matrix diagonalization, first and second order perturbation theory and confirmed experimentally. Magnetic field magnitude was measured by ^{13}C ENDOR signal with 0.02% precision or 0.5 mT. The orientation of quadrupole, hyperfine and fine structure tensors are the same within error of experiment, *g*-factor is isotropic.

1. Introduction

The study of NV^- centres has a long story from 1960-ies, firstly started from the optical characterisation [1] and then proceeds by EPR spectroscopy. Comparative analysis of the optical transitions and EPR lines was provided as early as 1970 in the work of Wyk [2], but correct interpretation of observed light-induced behaviour was done by [3], and relevant energy level scheme was proposed. The next major insight into the study of NV enters was made in the pioneering work [4] of the single-spin spectroscopy via NV centres. From that time, in the middle of 90-s, the NV centres was tried in many applications e.g. towards nanoscale system [5] as a highly efficient probe or qubit system [6]. Nowadays single nuclear spin spectroscopy was demonstrated even at room temperature [7,8] by means of coupling nuclear spin to the NV^- electronic spin, which is used to read-out nuclear spin state. The same recipe is used to prepare quantum qubit state using NV^- centre in diamond [9]. Therefore, to be reliable probe (or a qubit component) NV^- centre magnetic parameters, especially hyperfine and quadrupole tensors, have to be determined precisely. There are well-spread values of these spectroscopic parameters that are used routinely [10] in literature, but we haven't found a single work that determines *A* and *P* (hyperfine and quadrupole, correspondingly) on the solid basis of angle-dependent ENDOR spectroscopy. Moreover, there is a disagreement on the value and especially sign of *A* and *P* parameters. Thus, authors decided to fill this gap and provide results of their spectroscopic investigation of NV^- centre in particular commercial diamond by high-frequency ENDOR.

2. Materials and methods

Sample for the study was as used in work [11]. A micron-sized (of about 250 μm) diamond single crystal was fabricated commercially by Element Six using HPHT (high pressure high temperature) synthesis. The initial concentration of the nitrogen impurities in the sample was estimated to be of $\approx 5 * 10^{18}$ cm^{-3}. The crystal was subjected to electron irradiation (≈2 MeV) with the flux density of $\approx 10^{18}$ cm^{-2} followed by annealing in the hydrogen atmosphere at $T = 800$ °C for 2 h.

The experimental work of the paper was done on the Bruker Elexsys E680 machine operating at 94 GHz in the pulsed regime. Pulsed ENDOR, recorded by Mims pulse sequence is easier to implement and detect compared to the CW ENDOR (although there is a beautiful CW ENDOR study of P1 centre in diamond [12]), because the relaxation times (that usually have to be balanced with each other and temperature of the experiment) are directly involved to the Mims pulse sequence; in addition, high-frequency ENDOR spectrum has large frequency span and is easier to interpret. Field-swept EPR spectra were acquired using two-pulse Hahn echo sequence $\pi/2$–τ–π with $\pi/2$ pulse of 48 ns and $\tau = 280$ ns. ENDOR spectra were measured by Mims pulse sequence $\pi/2$–τ–$\pi/2$–T–$\pi/2$ with $\pi/2$ pulse of 48 ns (if other value is not mentioned) and $\tau = 280$ ns and RF pulse of 150 μs inserted in the time interval T

* Corresponding author.
E-mail address: boris.yavkin@gmail.com (B.V. Yavkin).

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