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NANOSTRUCTURE CHARACTERIZATION

Hyperfine Characteristics of Quantum Registers NV-^{13}C in Diamond Nanocrystals Formed by Seeding Approach from Isotopic Aza-Adamantane and Methyl-Aza-Adamanthane

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Abstract—We predict the characteristics of hyperfine interactions (*hfi*) for a number of electron-nuclear spin systems NV-^{13}C in diamonds grown by seeding approach from the specific isotopic aza-adamantane or methyl-aza-adamantane molecules differing in ^{13}C position in the precursor as well as in the orientation of the NV center in the post-obtained diamond. For the purpose we have used the spatial and *hfi* data simulated previously for the cluster $C_{510}[NV]^-H_{252}$. The data obtained can be used to identify (and correlate with the precursor used) the specific NV-^{13}C spin system by measuring the *hfi*-induced splitting in optically detected magnetic resonance spectra being characteristic for the NV-^{13}C system.

Keywords: nitrogen-vacancy (NV) color center, diamond, ^{13}C nuclear spin, precursor, aza-adamantane, methyl-aza-adamantane, hyperfine interaction, density functional theory

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INTRODUCTION

Nanostructured diamonds hosting optically active paramagnetic color centers (NV, SiV, GeV etc.) and hyperfine-coupled with them quantum memory ^{13}C nuclear spins located in some sites of a diamond lattice are currently of great interest to implement emerging second-generation quantum technologies (quantum information processing, quantum sensing and metrology). Current methods (high pressure high temperature (HPHT), chemical vapor deposition (CVD), fast ion beam implantation with subsequent annealing etc.) of creation such electronic-nuclear spin systems are inherently probabilistic with respect to mutual location of the color center electronic spin and the ^{13}C nuclear spins.

Recently new bottom-up approach to fabricate such systems was suggested (see, e.g. [1–4]) based on the idea to synthesize first chemically appropriate diamond-like organic molecules containing desired constituents in definite positions and then use them as precursors/seeds for CVD or HPHT growth to produce macroscopic diamonds. In particular, diamonds incorporating coupled NV-^{13}C spin systems (quantum registers) with definite mutual arrangements of NV and ^{13}C can thus be obtained from isotopically substituted aza-adamantane $C_9H_{15}N$ or methyl-aza-adamantane $C_{10}H_{17}N$ (see examples in Fig. 1). Very recently first successful experiments [5–7] have been done to implement this idea and macroscopic diamonds were grown from series of lower diamondoids using laser-heated diamond anvil cell.

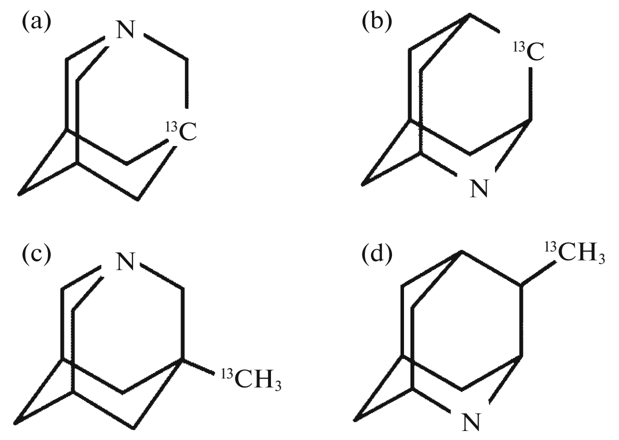

Fig. 1. Examples of (a) 1-aza-adamantane (4 equivalent sites in the molecule) and (b) 2-aza-adamantane (6 equivalent sites) with the isotopic ^{13}C atom located in different possible positions in the adamantane molecule and (c), (d) respective methyl-aza-adamantanes having ^{13}C atom in the methyl group.

BASIC RESULTS

Here we are presenting the simulated data which can be used to correlate the finally obtained NV-^{13}C spin system with the precursor used. For the purpose one can measure the hyperfine-induced splitting of the $m_S = \pm 1$ sublevels ($S = 1$ is the NV ground-state electronic spin) of the NV-^{13}C system using optically detected magnetic resonance (ODMR). As is well-known (see, e.g. [8]) this splitting is characteristic for NV-^{13}C systems having different location of the ^{13}C nuclear spin with respect to the NV center. Therefore, here we are predicting the characteristics of hyperfine interactions (*hfi*) for a number of NV-^{13}C systems in the diamonds grown from few specific isotopic aza-adamantane (Figs. 1a, 1b) and methyl-aza-adamantane (Figs. 1c, 1d) molecules differing in ^{13}C position in the precursor as well as in the orientation of the NV center in the post-obtained diamond.

As is well known (see e.g. [9]), the nitrogen atom in an aza-adamantane molecule can take the tertiary (nodal or bridgehead) positions (1-aza-adamantane, see examples in Figs. 1a, 1c) and secondary (bridge) positions (2-aza-adamantane, Figs. 1b, 1d).

In turn, after the growth of the macroscopic diamond from the precursors shown in Fig. 1, the vacancy can be formed by removing one of the C atom neaghboring the N atom of the precursor. Such vacancies can be located in four possible positions with respect to the N atom of the seed aza-adamantane molecule as it is shown in Fig. 2a for the 1-aza-adamantane (positions I–IV) and in Fig. 2b for the 2-aza-adamantane (positions V–VIII). Accordingly, hereinafter we will refer to the NV-^{13}C coupled spin systems resulting from the precursors a–d of Fig. 1 with the vacancy positions I–VIII near the nitrogen atom shown in Fig. 2 as aI, aII, aIII, aIV, bV, bVI, bVII, bVIII, cI, cII, cIII, cIV, dV, dVI, dVII and dVIII.

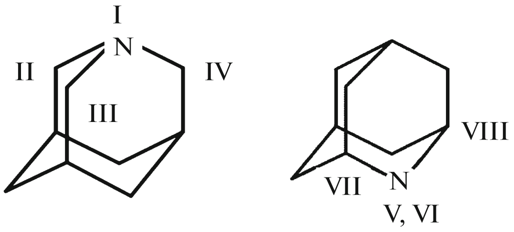

Fig. 2. Possible positions of the vacancy in the macroscopic diamond formed from 1-aza-adamantane (a) and 2-aza-admantane (b) precursors.

To model the macroscopic diamond crystal grown from the precursors shown in Fig. 1 and hosting the NV center with the vacancy in one of the position shown in Fig. 2 one can use a rather large relaxed H-terminated diamond cluster containing the NV center. Here we will use for the purpose the $C_{510}[NV]^-H_{252}$ cluster for which we previously [10] found spatial coordinates of all possible locations of the ^{13}C atom C(j) (j = 1–510) in the cluster with respect to the NV center and calculated full *hfi* matrices for all such NV-^{13}C spin systems in the cluster at specific orientation of the NV center in its central part (see [10] for details). Rotating this cluster so as to obtain the desired configuration of one of the considered isotopic precursors of Fig. 1 (from which this cluster can be grown), we were able to identify the number C(j) of the ^{13}C nuclear spins in this precursor. Then using table in the Supplement to the article [10], we found the *hfi* characteristics for the NV-^{13}C system obtained from the specific precursor as a result of diamond growth and creation a vacancy in one of the lattice site near the nitrogen atom.

The calculated data for the indicated positions (as they have been enumerated in [10] for the $C_{510}[NV]^-H_{252}$ cluster) of the isotopic ^{13}C carbon atom in different precursors are shown in Table 1. In the first column of Table 1 we indicate the precursor type according to Fig. 1 with indication of the vacancy position as shown in Fig. 2 and with the position number of the respective carbon ^{13}C atom in the $C_{510}[NV]^-H_{252}$ cluster. In the second, third and fourth columns of Table 1 we show the calculated values of the elements of the *hfi* matrix A_{ZZ}, $T_{nd} = \sqrt{A_{ZX}^2 + A_{ZY}^2}$ and A_{iso} = SpA/3 for respective NV-^{13}C spin system. The fifth column of Table 1 shows the calculated values of the *hfi*-induced splitting Δ_0 of the sublevels $m_S = \pm 1$ at zero external magnetic field, which can be measured experimentally and serve as the main parameter identifying the position of the ^{13}C nuclear spin in the diamond lattice relative to the NV center.

Table 1. Predicted values of the *hfi* parameters A_{ZZ}, T_{nd} and A_{iso}, as well as *hfi*-induced Δ_0 splitting of the $m_S = \pm 1$ sublevels of the NV-^{13}C system aI-dVIII presented in Figs. 1 and 2

Number of the ^{13}C position in the C_{510}[NV]-H_{252} cluster	A_{ZZ}, kHz	T_{nd}, kHz	A_{iso}, kHz	Δ_0, kHz
aI/C(417)	152.4	92.6	−73.7	178.3
aII/C(320)	3561.2	744.4	2990.3	3637
aIII/C(322)	3589.9	744.4	3016.3	3665.1
aIV/C(6)	136800	19767	152400	134000
bV/C(415)	344.9	150.2	45.5	376.3
bVI/C(343)	3691.1	742.1	3018.4	3665.8
bVII/C(331)	3580.5	744.1	3008.9	3655.8
bVIII/C(6)	136800	19767	152400	134000
cI/C(416)	168.1	131.4	50.2	213.4
cII/C(306)	−204.5	233.8	−233	310.6
cIII/C(308)	−203.3	233.8	−231.9	309.9
cIV/C(172)	−8484.4	827.4	−9557.4	8542.2
dV/C(412)	169	131.5	51.3	214.2
dV/C(414)	152.4	92.6	−73.7	178.3
dVI/C(257)	−6334	936	−5416	6406.5
dVI/C(340)	−201.5	233.3	−230.4	308.3
dVII/C318	−226.1	397.3	−441.8	457.1
dVII/C(329)	−203	233.4	−231.8	309.3
dVIII/C(259)	−6338.3	938.5	−5417.3	6406.5
dVIII/C(172)	−8484.4	827.4	−9557.4	8542.2

One can see that in the cases of aI, aII, aIII, aIV, bV, bVI, bVII, bVIII, cI, cII, cIII, cIV systems there are only one position of ^{13}C nuclear spin while for the dV, dVI, dVII and dVIII systems—two possible and different positions of the ^{13}C atom. Note also that the cases aIV/C(6) and bVIII/C(6), exhibiting largest *hfi*-induced splitting Δ_0, corresponds to the situation when the ^{13}C nuclear spin is located in the position C(6) being closest to the vacancy, in which the electron spin of the NV center interacts most strongly with the nearest-neighbor nuclear spins. The results obtained clearly demonstrate essentially different *hfi* characteristics (and, respectively, different *hfi*-induced splitting of the zero-field ODMR lines) for different NV-^{13}C systems thus providing the way to identify precursor and to determine position of the vacancy with respect to the N atom in the precursor.

In conclusion, we performed here the analysis of hyperfine interactions in macroscopic diamond nanoclusters obtained from four exemplary isotopically substituted aza-adamantane and methyl-aza-adamantane molecules which can be used as the precursors for growth of nanodiamonds. The values of the hyperfine characteristics calculated for various obtained NV-^{13}C complexes can be used to identify the type of precursor from which the corresponding diamond nanocluster with an NV center was obtained. Similar data can be obtained for other precursors, in particular, diadamantanes, triadamantanes, etc. Moreover, silicon or germanium can be substituted for nitrogen in them (see, e.g. [11]), because the corresponding silicon-vacancy and germanium-vacancy color centers are actively studied currently, since they are in some respects superior to the NV centers discussed here.

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CONFLICT OF INTEREST

The authors declare that they have no conflicts of interest.

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