SINGLE NV CENTERS IN NANOSTRUCTURED DIAMOND FOR QUANTUM INFORMATICS AND QUANTUM MAGNETOMETRY

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Abstract – Using density functional theory we have simulated hyperfine interactions (hfi) in the $C_{291}NVH_{172}$ cluster for all possible positions of the ^{13}C atom in the cluster and calculated hfi splittings of the EPR lines for arbitrary $^{14}NV+^{13}C$ spin systems. Applications of the systems for quantum information processing and single-spin quantum magnetometry will also be reviewed.

I. INTRODUCTION

The ability to create, control and measure the coherence in multi-spin systems in solids is crucial for scalable applications of quantum information processing, quantum sensing and metrology. Coupled electron-nuclear spin systems where electrons act as fast processing qubits while nuclei can store quantum information for a long time owing to their exceptional isolation from environment are especially useful for the purposes.

The most successful and promising representative of such systems is the nitrogen-vacancy (NV) color center in diamond [1] whose ground-state electron spin (e-spin) S=1 is coupled to the nuclear spin (n-spin) $I^{(N)}$=1 of its own ^{14}N atom and, potentially, to nearby n-spins $I^{(C)}$=1/2 of isotopic ^{13}C atoms that are distributed randomly in diamond lattice with the 1.1% probability. Hyperfine interactions (*hfi*) in such systems lead directly to a few-qubit gates which can be implemented using a sequence of optical, microwave or radio frequency pulses to initialize, coherently manipulate and readout the electron-nuclear spin system states [2-4]. Initially, it has been done [3] on single NV centers strongly coupled to a ^{13}C n-spin being nearest neighbor of the vacancy. Later [2,4] more distant ^{13}C nuclear spins located in the third coordination sphere have been distinguished in optically detected magnetic resonance (ODMR) spectra and spin echo modulation. Most recently usage of dynamical decoupling methods to suppress background spin noise allows to observe single NV centers coupled to much more distant single ^{13}C nuclear spins and to study them systematically [5-8]. Here, we report on the systematic study of hyperfine interactions between the electronic spin of single NV center and ^{13}C nuclear spins in the NV-hosting H-terminated carbon cluster $C_{291}NVH_{172}$ using computational chemistry simulation.

II. METHODS AND RESULTS

The geometric structure of the cluster was optimized and the spin density distribution was calculated by DFT using the B3LYP1 functional with the MINI/3-21G basis sets. The calculations have been performed for singly negatively charged cluster in the triplet ground state (S=1). We used the PC GAMESS (US) and ORCA software packages to calculate *hfi* matrices for all possible positions of the ^{13}C atom in the cluster. To be general, it has been done in the principle axis system (PAS) of the NV center where the Z axis coincides with the C_{3V} symmetry axis of the center while the X and Y axes are chosen arbitrarily. Evidently, various ^{13}C lattice sites showed different and generally anisotropic interactions with the NV e-spin, leading to different spin properties of various $NV+1^{13}C$ spin systems.

The simulated *hfi* matrices have been used in the standard spin Hamiltonian of an arbitrary $^{14}NV+1^{13}C$ system that took into account i) zero-field fine structure splitting of the ^3A ground-state of the center in a diamond crystal field, ii) *hfi* of the S=1 e-spin of the NV center with I=1 n-spin of the ^{14}N atom of the center, iii) the quadrupole moment Q=1 of the ^{14}N nucleus, iv) *hfi* with the I=1/2 n-spin of a ^{13}C nucleus disposed somewhere in the cluster and v) Zeeman interactions of all three spins with arbitrarily directed external magnetic field. Numerical diagonalization of these spin Hamiltonians

provides 18 eigenenergies and respective 18 eigenstates of all possible ^{14}NV+1^{13}C spin systems in the cluster.

Using this approach we have simulated spectra of optically detected magnetic resonance (ODMR) of ^{14}NV+1^{13}C spin systems and compare them with those experimentally observed in [6]. Typical *hfi* structure of e.g. $m_S=0 \leftrightarrow m_S=-1$ line in the ODMR spectrum of a system in low magnetic field consisted of six lines corresponding to allowed EPR transitions in the system with their frequency differences determined by the *hfi* with the ^{14}N and ^{13}C nuclear spins. From these ODMR spectra one can extracts zero-field splittings Δ_{0i} of e.g. the $m_S=-1$ NV e-spin state resulted from its *hfi* with single ^{13}C n-spin taking specific (i-th) position in diamond lattice with respect to the NV center. If we compare these experimental data with those obtained by spin-Hamiltonian method using simulated *hfi* matrices for all possible ^{14}NV+^{13}C system we will be able to address the specific ^{13}C nucleus among other positions.

Simulated values of hfi and spatial characteristics for 121 positions of a ^{13}C n-spin in the $C_{291}NVH_{172}$ cluster are presented in the Table 1. Calculations showed that owing to the C_{3V} symmetry of the NV center there are N_C (=3 or 6) positions of ^{13}C nuclei in the cluster exhibiting very close values of their hfi and spatial characteristics. In the Table 1, we indicate data for 26 sets of such near-equivalent lattice sites (families) which are termed by English alphabet letters A-Z with indication of most important elements A_{ZZ}, $A_{nd} = (A_{XZ}^2 + A_{YZ}^2)^{1/2}$ of hfi matrices, zero-field *hfi* splittings Δ_{0i}, Z coordinates, distances from Z-axis and from N atom of the NV center. All data are averaged over the family members.

TABLE 1 – Simulated *hfi* and spatial characteristics for the "families" of ^{13}C n-spin in the $C_{291}NVH_{172}$ cluster.

Family	N_C	$\overline{A}_{ZZ}$ (MHz)	$\overline{A}_{nd}$ (MHz)	$\overline{\Delta}_i^{(0)}$ (MHz)	$\overline{Z}$ (Å)	$\overline{r}_{XY}$ (Å)	$\overline{r}_{NC}$ (Å)
A	6	12.451	1.166	12.471	- 0.522	3.937	4.536
B	3	11.386	1.434	11.451	- 2.655	2.972	5.298
C	3	-8.379	0.827	8.437	- 2.109	1.487	4.118
D	6	-6.450	0.931	6.521	- 0.010	2.552	3.089
E	3	4.055	0.826	4.136	-2.643	1.491	4.621
F	6	3.609	0.738	3.682	1.577	2.562	2.566
G	6	2.281	0.240	2.292	0.008	5.166	5.446
H	3	1.884	0.208	1.895	-4.242	2.976	6.673
I	3	-1.386	0.130	1.392	0.005	4.458	4.780
J	6	-1.145	0.328	1.191	-2.110	3.932	5.497
K1	3	-0.886	0.510	1.022	-2.118	2.985	4.871
K2	3	-1.011	0.014	1.012	-0.002	4.460	4.785
L	3	0.980	0.121	0.986	-0.535	2.972	3.737
M	3	0.602	0.557	0.819	2.127	1.460	1.513
N	6	0.725	0.095	0.731	-0.541	6.467	6.855
O1	3	0.673	0.171	0.694	-4.712	4.479	7.847
O2	3	0.655	0.166	0.676	3.707	2.983	3.578
P	6	0.474	0.190	0.510	-2.635	5.355	6.909
Q	6	0.391	0.273	0.477	-2.645	3.953	5.897
R	3	-0.226	0.393	0.453	2.115	2.985	3.009
S	3	0.412	0.060	0.417	-0.511	5.942	6.351
T	3	0.366	0.149	0.395	3.709	1.504	2.485
U	3	0.286	0.225	0.364	1.578	4.481	4.484
V	6	-0.209	0.232	0.312	2.105	3.927	3.945
W	3	-0.200	0.171	0.266	-4.220	1.489	6.135
X	6	0.211	0.152	0.259	-4.768	2.573	6.990
Y	6	-0.228	0.001	0.227	-0.522	5.381	5.834
Z1	3	0.158	0.131	0.205	4.226	3.001	3.903
Z2	3	0.086	0.184	0.203	1.576	4.447	4.447
on-NV-axis	1	0.187	0.001	0.187	-4.734	0.009	6.465

we do not show in the Table 1 largest simulated *hfi* splittings of ~130 MHz for the three sites being NN of the vacancy, as they are well documented in the literature.

We have shown that all simulated data correlate well with available experimental data of works [5-8] which demonstrates that *hfi* parameters simulated for the $C_{291}NVH_{172}$ cluster by DFT in conjunction with spin Hamiltonian method provide good fit to the experimental *hfi* splittings, allowing simultaneously to address possible positions of ^{13}C in diamond lattice. Moreover, we were able to describe well the experimental ODMR spectra shown in [6] for the specific $^{14}NV+^{13}C$ spin system. We also have calculated cosines of angles between Z axis of the NV PAS and z axis of ^{13}C PAS for all possible positions of ^{13}C in the cluster. For a first time we predict the zero-field *hfi* splitting of 187.4 kHz for the nearest ^{13}C atom position lying on the NV axis. These data will be published elsewhere.

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