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DIAMOND SIDE RESONANCES: INFLUENCE OF ISOTOPIC SUBSTITUTION OF CARBON

ALEXANDER A. IVANOV AND ALEXEI I. IVANOV

Immanuel Kant Baltic Federal University
Nevskogo 14, 236004 Kaliningrad, Russia

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Abstract: We discuss the side resonances of the optically detected magnetic resonance in a diamond crystal and propose a new approach to the calculation of the hyperfine interaction in a composed system consisting of a negatively charged nitrogen-vacancy NV^- center and a nearby ^{13}C nuclear spin. The energy levels, rule selection and radiative transitions are obtained by a new method. The base of this method is the use of a complete set of commuting operators and entangled spin states. An estimation of the carbon hyperfine splitting parameters in the diamond NV^- center from side-resonance frequencies is obtained in the frame of this method.

Keywords: diamond, hyperfine structure, side resonances

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1. Introduction

Control over individual electronic and nuclear spins in the solid state offers a promise for applications in quantum information processing (QIP) and field sensing at a nanometer scale. The negatively charged nitrogen-vacancy NV^- center in diamond has recently emerged as a candidate for QIP and magnetometry [1]. The optical transitions of the NV^- center allow a high degree of spin polarization at room temperature via optical pumping. By optically pumping with a laser light in the green spectral range the NV^- center is initialized into $m_s = 0$ of its triplet ground state. Optically detected magnetic resonance (ODMR) signals enable probing the energy levels and spin states of NV^- centers.

In the absence of magnetic fields, the NV^- center has a magnetic resonance at a frequency of approximately 2870 MHz, which corresponds to a transition between the triplet ground-state magnetic sublevels $m_s = 0$ and $m_s = \pm 1$. Side-resonances around this central resonance have been reported in the literature [2]. In ref. [2] the asymmetrically displaced side-resonances separated by around 130 MHz were attributed to hyperfine interaction of the NV^- center with a nearest-neighbor ^{13}C nuclear spin.

In this paper an estimation method of the carbon hyperfine splitting parameters from side-resonances frequencies is proposed. The energy levels, rule selection and radiative transitions are obtained by a new approach. The base of this approach is the use of a complete set of commuting operators and entangled spin states. This approach was used by us earlier in the calculation of nuclear magnetic resonance spectra [3].

2. Method and results

The negatively charged NV^- center in diamond consists of a substitutional nitrogen atom associated with a vacancy in an adjacent lattice site of the diamond matrix. Its ground state is a spin triplet ($S=1$) with an intrinsic spin quantization axis provided by the NV^- center symmetry axis. We consider a single NV^- center associated with native ^{14}N isotopes (99.6% abundance), corresponding to a nuclear spin $I=1$. Hyperfine coupling of the electron spin of the NV^- center occurs to ^{13}C (1.1% abundance) in the surrounding lattice and to ^{14}N at the center. The hyperfine coupling of nearest neighbor carbons (nuclear spin $I=1/2$) is around 130 MHz [4]. The hyperfine coupling of the electron spin to ^{14}N at the NV^- center is around 3 MHz [1]. The ground-state spin Hamiltonian in frequency units reads as

$$\hat{H}=D\left(\hat{S}_z^2-\frac{\hat{S}^2}{3}\right)+A^{\parallel}\hat{S}_z\hat{I}_z+A^{\perp}(\hat{S}_x\hat{I}_x+\hat{S}_y\hat{I}_y) \tag{1}$$

where $D\approx 2870$ MHz is the fine structure splitting, $A^{\parallel}$ and $A^{\perp}$ are the axial and non-axial magnetic hyperfine parameters of the ^{13}C nucleus, the z-axis coincides with the electronic spin quantization axis.

To calculate the spectrum of the Hamiltonian we use the method that we have proposed in ref. [3]. For this purpose, the total spin operator first determines

$$\hat{\vec{J}}=\hat{\vec{S}}+\hat{\vec{I}} \tag{2}$$

and, based on the principles of the quantum theory of angular momentum, it will build vectors $|J,\, M_z\rangle$ which are eigenvectors of a complete set of the commuting operators $\hat{J}^2$, $\hat{J}_z$, $\hat{S}^2$, $\hat{I}^2$:

$$\begin{gathered}\left|\tfrac{3}{2},\tfrac{3}{2}\right\rangle=|1,1\rangle\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle\\ \left|\tfrac{3}{2},\tfrac{1}{2}\right\rangle=\sqrt{\tfrac{2}{3}}|1,0\rangle\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle+\tfrac{1}{\sqrt{3}}|1,1\rangle\left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle\\ \left|\tfrac{3}{2},-\tfrac{1}{2}\right\rangle=\sqrt{\tfrac{2}{3}}|1,0\rangle\left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle+\tfrac{1}{\sqrt{3}}|1,-1\rangle\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle\\ \left|\tfrac{3}{2},-\tfrac{3}{2}\right\rangle=|1,-1\rangle\left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle\end{gathered} \tag{3}$$

$$\begin{gathered}\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle=\sqrt{\tfrac{2}{3}}|1,1\rangle\left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle-\tfrac{1}{\sqrt{3}}|1,0\rangle\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle\\ \left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle=\tfrac{1}{\sqrt{3}}|1,0\rangle\left|\tfrac{1}{2},-\tfrac{1}{2}\right\rangle-\sqrt{\tfrac{2}{3}}|1,-1\rangle\left|\tfrac{1}{2},\tfrac{1}{2}\right\rangle\end{gathered} \tag{4}$$

Despite the fact that in (3)–(4) the vectors of admissible states are classified by multiplets, we note that the total spin J is not conserved, as the Hamiltonian $\hat{H}$

form (1) does not commute with the operator $\hat{J}^2$. At the same time, the Hamiltonian commutes with the projection of the total spin, the square of the electron spin and the nuclear spin square:

$$\left[\hat{H},\hat{J}_z\right]=\left[\hat{H},\hat{S}^2\right]=\left[\hat{H},\hat{I}^2\right]=0 \tag{5}$$

The set of operators $\hat{H}$, $\hat{J}_z$, $\hat{S}^2$, $\hat{I}^2$ is also a complete set of commuting operators. This set of operators has a unique system of eigenvectors. Consequently, the vectors of the NV^- center ground state are characterized by the value of energy E, the projection of the total spin M_z, the electron spin S and the nuclear spin I. Since for all these states $S=1$, $I=1/2$, then the equation for the eigenvalues and eigenvectors of the Hamiltonian (1) can be written as

$$\hat{H}\left|E^{(i)}_{M_z}\right\rangle = E^{(i)}_{M_z}\left|E^{(i)}_{M_z}\right\rangle \tag{6}$$

Where the index i is introduced in order to distinguish the states with the same value M_z and different values of energy E.

First, we note that the vectors $\left|E_{\pm\frac{3}{2}}\right\rangle=\left|\frac{3}{2},\pm\frac{3}{2}\right\rangle$ satisfy the equation (6) with the eigenvalue

$$E_{\pm\frac{3}{2}}=\frac{D}{3}+\frac{A^{\parallel}}{2} \tag{7}$$

Secondly, since the total spin is not conserved, the stationary states with $M_z=\pm 1/2$ will be represented as superpositions

$$\left|E^{(i)}_{\pm\frac{1}{2}}\right\rangle = c_1^{(i)}\left|\frac{3}{2},\pm\frac{1}{2}\right\rangle + c_2^{(i)}\left|\frac{1}{2},\pm\frac{1}{2}\right\rangle \tag{8}$$

Substituting (8) into (6) and solving the resulting equation, we find the energy levels

$$E^{(1)}_{\pm\frac{1}{2}}=-\frac{2D}{3}+V+P \quad E^{(2)}_{\pm\frac{1}{2}}=-\frac{2D}{3}+V-P \tag{9}$$

where $V=\frac{D}{2}-\frac{A^{\parallel}}{4}$, $P=\sqrt{V^2+\frac{(A^{\perp})^2}{2}}$.

Thus, for the Hamiltonian (1), taking into account the laws of conservation (5) two groups of stationary states are found. Indeed, the first group (with energies of about $\frac{D}{3}$) includes the states $\left|E_{\pm\frac{3}{2}}\right\rangle$ and $\left|E^{(1)}_{\pm\frac{1}{2}}\right\rangle$. The second group (with energies of about $\frac{-2D}{3}$) includes the states $\left|E^{(2)}_{\pm\frac{1}{2}}\right\rangle$. Radiative transitions between these states are subject to the selection rule $\Delta M_z=\pm 1$. The main peak of ODMR of the NV^- center occurs at a frequency of $\nu_0=2870$ MHz.

Side resonances correspond to transitions $\left|E^{(2)}_{\frac{1}{2}}\right\rangle \leftrightarrow \left|E^{(1)}_{-\frac{1}{2}}\right\rangle$, $\left|E^{(2)}_{-\frac{1}{2}}\right\rangle \leftrightarrow \left|E^{(1)}_{\frac{1}{2}}\right\rangle$ with frequency ν_h and transitions $\left|E^{(2)}_{\frac{1}{2}}\right\rangle \leftrightarrow \left|E_{\frac{3}{2}}\right\rangle$, $\left|E^{(2)}_{-\frac{1}{2}}\right\rangle \leftrightarrow \left|E_{-\frac{3}{2}}\right\rangle$ with frequency ν_l, respectively. Thus, the hyperfine interaction anisotropy is shown in the structure of side resonances in diamond. Moreover, if $A^{\parallel}$ and $A^{\perp}$ are not equal, these side resonances are located asymmetrically relatively to the center.

According to [2], the distance between the non-symmetrical resonances is significant and equal to $\Delta_C \simeq 126(2)$ MHz. The reason for the formation of resonances of this group is the interaction between the NV^- center and the ^{13}C nucleus.

If, based on this experimental data, take $\nu_0 - \nu_l = 56$ MHz and $\nu_h - \nu_0 = 70$ MHz, then for the axial and non-axial hyperfine splitting parameters, we obtain the following values: $A^{\parallel} = -121$ MHz and $A^{\perp} = \pm 166$ MHz. The authors of ref. [5] used the interaction of the electron spin NV^- center with the nearest ^{13}C nuclear spin for the demonstration of the quantum NOT gate and a conditional two-qubit gate. They used states of the form $|m_s\rangle|m_i\rangle$ with $m_s = \pm 1$, $m_i = \pm\frac{1}{2}$. Note that we have obtained entangled states (8) to become of the form $|m_s\rangle|m_i\rangle$ in ref. [5] only at the zero value of the non-axial hyperfine splitting parameter $A^{\perp}$. In this case, two side resonances were placed symmetrically relative to the central resonance. This arrangement was not observed in the experiment [2].

3. Summary

In conclusion we have studied the side resonances of the optically detected magnetic resonance in diamond crystal and propose a new approach to the calculation of the hyperfine interaction in a composed system, consisting of a NV^- center and a nearby ^{13}C nucleus. The energy levels, rule selection and radiative transitions are obtained by a new method. The base of this method is the use of a complete set of commuting operators and entangled spin states of the electronic spin of the NV^- center and the nearby ^{13}C nuclear spin. An estimation of the carbon hyperfine splitting parameters in the diamond NV^- center from side-resonance frequencies is obtained in the frame of this method.

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