

Theory of optical Λ transitions in the $^{15}\text{N-V}^-$ center in diamond: Role of the hyperfine interaction

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We present a thorough derivation of the excited-state energy levels of the negatively charged $^{15}\text{N-V}^-$ center in diamond for the strong applied electric-field case. We show that in the $^{15}\text{N-V}^-$ center a spin nonconserving two-photon Λ transition exists that is mediated by the hyperfine interaction, which provides the possibility to write quantum information. Using second-order perturbation theory we obtain a Λ transition rate of the order of 10 MHz at room temperature, which allows for approximately 10^4 quantum logic operations within the spin coherence time $\tau_d(T=300\text{ K}) \approx 1\text{ ms}$ of the $^{15}\text{N-V}^-$ center.

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Recently tremendous attention has been drawn by single nitrogen-vacancy defect centers (N-V centers) in diamond due to their promising properties for the experimental realization of quantum computation.¹ In particular, the N-V center in diamond is well suited for studying electronic and nuclear-spin phenomena since its spin can be both initialized and read out via a stable optical spin conserving transitions.^{2,3} The N-V center appears in two forms, neutral and negatively charged, and it is the negatively charged center (N-V⁻) that we are interested with in this rapid communication. The N-V⁻ stands out among solid-state systems because its electronic spin can be efficiently prepared, manipulated and measured with optical and microwave excitations. Recent experiments have conclusively demonstrated that the N-V center in ultrapure diamond lattice shows the longest room-temperature spin dephasing time ever observed in solid-state systems ($T_2=1.8\text{ ms}$).⁴ The electron-spin coherence lifetime in the N-V⁻ center is limited by its hyperfine interaction with the Carbon lattice in the diamond structure.⁵ At very low concentration of paramagnetic impurities the loss of electron-spin coherence is minimized and spin quantum effects could be observed at room temperature over hundred of nanometers, these results open the door to coherent manipulation of individual isolated nuclear spins in a solid-state environment at room temperature with potential applications to quantum computation, therefore the understanding of the hyperfine interaction in this solid-state system is of great interest.

In this rapid communication we show how the hyperfine interaction gives rise to a spin nonconserving two-photon Λ transition with a rate of the order of 10 MHz at room temperature, which allows for approximately 10^4 optically controlled quantum logic operations within the spin coherence time at room temperature. The Λ transition can be understood by considering the hyperfine interaction of the electron's spin in the N-V⁻ center with the spin-1/2 nucleus of the isotope Nitrogen ^{15}N . A quantitative result for the number of quantum logic operations is obtained by using second-order perturbation theory and numerical diagonalization of the Hamiltonian describing the excited energy levels of the $^{15}\text{N-V}^-$ center when an external electric field is applied. Our results clarify the origin of the two-photon transitions that

have been experimentally observed in Refs. 6 and 7.

The N-V center in diamond consists of a substitutional nitrogen atom next to a Carbon vacancy giving a centre with C_{3v} symmetry. In the negatively charged state, the extra electron is located at the vacancy site forming a spin $S=1$ pair with one of the vacancy electrons. The energy-level structure of the N-V⁻ center was established by combining optical electron-paramagnetic resonance and theoretical results. Leneff and Rand earlier work gave a group theoretical description of the molecular orbitals and state configuration of the N-V⁻ center by using linear combinations of sp^3 orbitals.⁸ Recent numerical *ab initio* calculations conducted by Hossain *et al.* indicated the following orthonormal orbitals which irreducibly transform according to the C_{3v} symmetry group: $u=s_N$, $v=(s_1+s_2+s_3-3\beta s_N)/\sqrt{3}S_1$, $e_x=(2s_3-s_1-s_2)/\sqrt{3}S_2$, $e_y=(s_1-s_2)/S_2$, where s_N , s_1 , s_2 , and s_3 denote sp^3 orbitals pointing to the Nitrogen and three Carbons neighboring the vacancy, $S_1=\sqrt{1+2\alpha-3\beta^2}$, $S_2=\sqrt{2-2\alpha}$, $\alpha=\langle s_1|s_2\rangle$ and $\beta=\langle s_3|u\rangle$ are normalization constants.⁹ The energy levels are traditionally labeled according to the orbital and spin multiplicity, which is related to the number of states having the same energy. For the negatively charged N-V center, the ground state is a spin triplet state 3A_2 , with a zero-field splitting of $D_{gs}=2.87\text{ GHz}$ between spin sublevels $m_s=0$ and $m_s=\pm 1$ (see Fig. 1).

The excited-state 3E is also a spin triplet, associated with a broadband photoluminescence emission with zero-phonon line at 1.945 eV, which allows optical detection of single N-V defects using confocal microscopy. It is now well estab-

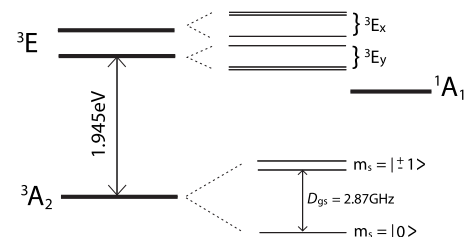


FIG. 1. Schematic diagram for the energy levels of an NV⁻ center depicting the spin triplets for the ground state 3A_2 and the excited-state 3E . The splitting due to spin-orbit interaction is shown for the excited state.

lished that a metastable state 1A_1 is placed between the ground and excited triplet states.¹⁰ Although an extensive documentation has been made to unveil the excited-state structure of the N-V center from first principles, it remains a topic of current research due to its possible quantum applications.¹¹

The Hamiltonian suitable to describe the 3E excited state of the negatively charged $^{15}\text{N-V}^-$ center is given by¹²

$$\mathcal{H} = H_{el} + \lambda \mathbf{L} \cdot \mathbf{S} + \rho \left[(\mathbf{L} \cdot \mathbf{S})^2 + \frac{1}{2} \mathbf{L} \cdot \mathbf{S} - \frac{1}{3} L(L+1)S(S+1) \right] + A \mathbf{I} \cdot \mathbf{S}. \quad (1)$$

The first term in Eq. (1) is the dominant term in the Hamiltonian and is given by $H_{el} = \mathbf{F} \cdot \mathbf{D}$, where \mathbf{F} is the external

applied electric field in the N-V defect and \mathbf{D} is the dipole moment operator, respectively. From earlier work done by Hossain *et al.* we know that the only nonzero dipole moment components are \mathbf{D}_{e_x} and \mathbf{D}_{e_y} with equal dipole magnitudes, i.e., $|\mathbf{D}_{e_x}| = |\mathbf{D}_{e_y}| = D$, respectively. The dipole moment transverse terms cause a linear splitting of the degenerate 3E state levels given by $\pm D \sqrt{(F_x + f_x)^2 + (F_y + f_y)^2}$,¹² where we have taken into account that the presence of strain has the same effect as a weak electric field denoted by \mathbf{f} . The second and third terms in Eq. (1) describe the spin-orbit and the spin-spin interaction in the LS coupling approximation,¹³ respectively.

With this information and restricting ourselves to the $m_l = \pm 1$ state sublevels of the spin triplet we can write the electric field, the spin-orbit and spin-spin interactions in the following matrix form

$$H_{el} + H_{ss} + H_{so} = \begin{pmatrix} -\lambda + \frac{13}{6}\rho & iD(f_y + F_y) & 0 & 0 & 0 & 0 & \rho \\ -iD(f_y + F_y) & \lambda + \frac{13}{6}\rho & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\frac{1}{3}\rho & iD(f_y + F_y) & 0 & 0 & 0 \\ 0 & 0 & -iD(f_y + F_y) & -\frac{1}{3}\rho & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \lambda + \frac{13}{6}\rho & iD(f_y + F_y) & 0 \\ \rho & 0 & 0 & 0 & 0 & -iD(f_y + F_y) & -\lambda + \frac{13}{6}\rho \end{pmatrix} \otimes \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}, \quad (2)$$

where we have chosen the basis $|m_l m_s\rangle$ in the following order $[|-11\rangle, |-1-1\rangle, |-10\rangle, |10\rangle, |11\rangle, |1-1\rangle]$ for numerical purposes and we have assumed that the electric field is applied in the y direction. The direct product between matrices takes into account the nuclear-spin subspace, i.e., $m_l = \pm 1/2$.

One can see that the spin-orbit interaction does not couple terms with different orbital projections and produces just an energy splitting between the states $m_s = \pm 1$. The spin-spin interaction however couples the first and the last state with different orbital projections and produces an energy splitting between the states $m_l = \pm 1$. The Hamiltonian given in Eq. (2) can be solved exactly and it is easy to convince oneself from the exact solution that there is no overlapping between eigenstates with different spin projections. Hence, the spin-orbit and spin-spin interaction cannot account for the mixing of the excited states and therefore cannot give rise to spin nonconserving transitions. If we want to allow for spin nonconserving transitions between one ground state and another via a common excited state we must introduce the nuclear-spin interaction. The hyperfine interaction will mix the eigenstates with different spin projections allowing us to have a transition to two different ground states. This particular transition is a second-order process that can be calculated by using Fermi's golden rule and is commonly known as a Λ transition. Therefore, the last term in the total Hamiltonian given in Eq. (1) is the nuclear-spin interaction and this term will be responsible for the mixing of the excited states, which will give rise to Λ transitions in the $^{15}\text{N-V}^-$ center in

diamond. The hyperfine interaction can be written in the above basis as

$$H_{ns} = A \begin{pmatrix} \frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -\frac{1}{2} & 0 & 0 & \sqrt{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -\frac{1}{2} & 0 & 0 & \sqrt{2} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & \sqrt{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & \sqrt{2} & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{2} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{2} & 0 & 0 & -\frac{1}{2} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & \sqrt{2} & 0 & 0 & -\frac{1}{2} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & \frac{1}{2} \end{pmatrix}, \quad (3)$$

where A is the excited-state hyperfine coupling constant which for the ^{15}N nuclear-spin case is $A \approx 60$ MHz.¹⁴ The eigenvalues of the total Hamiltonian given by Eq. (1) are plotted as a function of the electric field in Fig. 2. The energy diagram in Fig. 2 shows the mixing between the states $m_s = \pm 1$. This mixing will allow us to induce Λ transitions be-

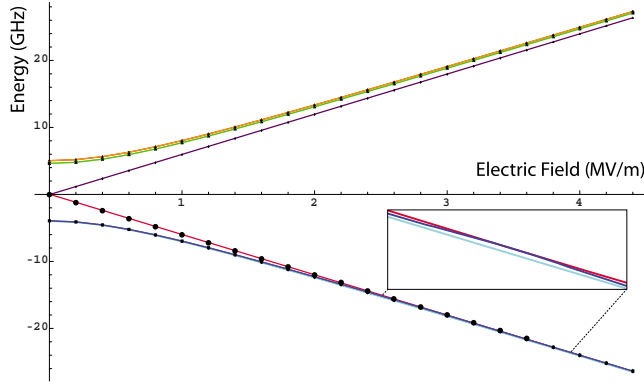


FIG. 2. (Color online) Energy diagram showing the eigenvalues of the total Hamiltonian given in Eq. (1) as a function of the applied external electric field. The lower branch shows the mixing between the states $m_s = \pm 1$. The eigenvalues are doubly degenerate.

tween these states in order to write quantum information by using circularly polarized light. The $^{15}\text{N-V}^-$ center has optical transitions between the ground state 3A_2 and the excited-state 3E at zero magnetic field that are spin conserving and will allow the readout of information through the electron's spin.¹² In order to write information we need to have optical transitions from two ground-state levels via a common excited state. The induced transition probabilities for the case of circularly polarized light around the z -axis is denoted by σ^\pm and is proportional to the modulus squared of the matrix element given by $\langle \psi_i | \sigma^\pm | \psi_j \rangle$, where ψ_i and ψ_j are electronic wave functions for the initial and final state, respectively, and $\sigma^\pm = \mp \sum_n (x_n \pm iy_n) / \sqrt{2}$, where the summation are over the electrons in the N-V^- center.

Suppose that in the absence of light the $^{15}\text{N-V}^-$ system is prepared in the ground state $|m_l m_s\rangle |m_I\rangle = |00\rangle |\uparrow\rangle$. If a strong external electric field with magnitude F_0 is applied and the crossing between different states occur at a given energy ϵ_n for the n^{th} excited state as illustrated in Fig. 2, then the ground state will be coupled to an excited state given by

$$|\Psi_n\rangle = \sum_{m_l = \pm 1, m_s = 0, \pm 1, m_I = \uparrow, \downarrow} C_{m_l m_s m_I}^n |m_l m_s m_I\rangle. \quad (4)$$

The coherent superposition of states given in Eq. (4) allows the possibility to have optical transitions between two ground states with different spin projections, i.e. spin non-conserving transitions, by using either a right or left circularly polarized light. In order for the final total angular momentum to equal the initial angular momentum, the nuclear spin projection flips [see Fig. 3]. The amplitude \mathcal{A}_\pm for the Λ transition in the dipole approximation from the initial state $|00\rangle |\uparrow\rangle$ to the final state $|01\rangle |\downarrow\rangle$ through either right or left circularly polarized light is given in second-order perturbation theory by

$$\mathcal{A}_\pm = e^2 \mathcal{E}^2 \sum_{n=1}^{12} \frac{\langle \downarrow | \langle 01 | \sigma^\pm | \Psi_n \rangle \langle \Psi_n | \sigma^\pm | 00 \rangle | \uparrow \rangle}{\omega_{\sigma^\pm} - (\epsilon_n - \epsilon_0)}, \quad (5)$$

where e is the electron charge, \mathcal{E} is the amplitude of the drive optical field, ϵ_0 is the energy of the initial ground state and

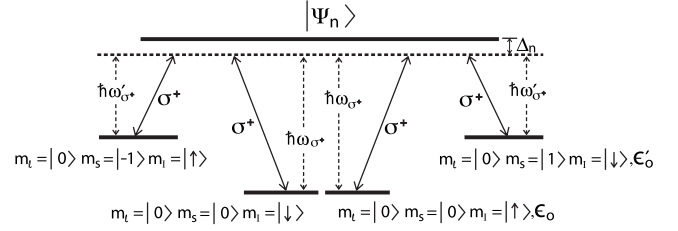


FIG. 3. The Λ transition optically driven with σ^\pm -polarized laser beams will flip the nuclear-spin projection in order for total angular momentum to be conserved. The Λ transition will equally populate both ground-state levels due to the twofold degeneracy between them.

ω_{σ^\pm} is the frequency of the circularly right or left polarized light, respectively. Writing the explicit form of $|\Psi_n\rangle$ in terms of the basis vectors in Eq. (5) we get

$$\mathcal{A}_\pm = e^2 \mathcal{E}^2 \sum_{n=1}^{12} \frac{C_{\mp 11 \downarrow}^n \sigma_{0, \mp 1}^\pm C_{\pm 10 \uparrow}^{n*} \sigma_{\pm 1, 0}^\pm}{\omega_{\sigma^\pm} - (\epsilon_n - \epsilon_0)}, \quad (6)$$

where $\langle i | \sigma^\pm | j \rangle = \sigma_{i,j}^\pm$. The value of the $C_{m_l m_s m_I}^n$ coefficients and eigenvalues ϵ_n can be obtained by numerical diagonalization of the total Hamiltonian for a given value of the applied external electric field F_0 (see Table I). In the subsequent calculations we will work with right circularly polarized only. In order to calculate the transition rate between the initial ground state and the final ground state we use Fermi's golden rule

$$W_{|00\rangle |\uparrow\rangle \rightarrow |01\rangle |\downarrow\rangle} = \frac{2\pi}{\hbar^4} |\mathcal{A}|^2 \rho(\omega), \quad (7)$$

where $\rho(\omega)$ represents the allowed photon frequencies and can be written in its simplest form as a Lorentzian function

TABLE I. Numerical values for the coefficients $C_{m_l m_s m_I}^n$ and eigenvalues ϵ_n of the excited state levels that contribute to the transition amplitude \mathcal{A}_\pm for the case $F_0 = 1.85$ MV/m (Ref. 15).

n	ϵ_n (GHz)	$C_{-11\downarrow}^n$	$C_{-10\uparrow}^n$	$C_{10\uparrow}^n$	$C_{11\downarrow}^n$
1	12.8540	0.5633	0.0056	$-i0.0021$	$i0.3965$
2	12.8540	-0.1011	-0.0010	$-i0.0004$	$-i0.0712$
3	12.4179	0.5754	0.0292	$-i0.0285$	$-i0.3770$
4	12.4179	-0.1540	-0.0078	$-i0.0076$	$i0.1009$
5	-11.3719	-0.1261	0.08755	$i0.0879$	$-i0.2099$
6	-11.3719	-0.3276	0.2274	$i0.2282$	$-i0.5452$
7	-11.1861	-0.2090	-0.6498	$-i0.6502$	$-i0.1005$
8	-11.1861	-0.0050	-0.0157	$-i0.0157$	$-i0.0024$
9	-11.1448	-0.3864	0.1344	$i0.1306$	$i0.5767$
10	-11.1448	0.0243	-0.0084	$-i0.0082$	$-i0.0362$
11	10.9909	0.0047	-0.1201	$i0.1201$	$-i0.0021$
12	10.9909	-0.0271	-0.6961	$-i0.6962$	$i0.0122$

$$\rho(\omega) = \frac{(\gamma/2\pi)}{[\omega_{\sigma^+} - \omega'_{\sigma^+} - (\epsilon'_0 - \epsilon_0)]^2 + (\gamma/2)^2}, \quad (8)$$

where ϵ'_0 is the energy of the final ground state and γ is the decoherence rate. For the resonant case, i.e. $\omega_{\sigma^+} - \omega'_{\sigma^+} = (\epsilon'_0 - \epsilon_0)$, Fermi's golden rule is given by $W_{|00\rangle|\uparrow\rangle \rightarrow |01\rangle|\downarrow\rangle} = 4|A|^2 / \gamma \hbar^4$. Taking the value of the dipole moment matrix elements as $e\sigma_{0,-1}^+ = e\sigma_{1,0}^+ = |d|$ in Eq. (6) we get

$$A = \hbar^2 \Omega_R^2 \sum_{n=1}^{12} \frac{C_{-11\downarrow}^n C_{10\uparrow}^{n*}}{\Delta_n}, \quad (9)$$

where $\Delta_n = \omega_{\sigma^+} - (\epsilon_n - \epsilon_0)$ is the detuning and we have used the relation $\mathcal{E}|d| = \hbar \Omega_R$ where Ω_R is the Rabi frequency. In order for perturbation theory to be valid we require that $|\Delta_n| \gg \Omega_R |C_{10\uparrow}^{n*}|$, hence we have to tune the frequency ω_{σ^+} to achieve this. Assuming that the detuning is taken in such a way that $|\Delta_n| \approx 30 \Omega_R |C_{10\uparrow}^{n*}|$, where the Rabi frequency is a few GHz,¹⁶ then the amplitude is $|A|^2 \approx 10^{-27} \hbar^4 \Omega_R^4$. Using the spin coherence time of $\gamma^{-1} \approx 1$ ms,⁴ we can estimate the Λ transition rate to be of the order of

$$W_{|00\rangle|\uparrow\rangle \rightarrow |01\rangle|\downarrow\rangle} \approx 10 \text{ MHz}. \quad (10)$$

Based on the obtained transition rate and for coherence lifetimes of around the milliseconds,⁴ we estimate that the $^{15}\text{N-V}^-$ center in diamond should be capable of around 10^4 quantum operations per coherence lifetime at room temperature, which confirms that this solid-state system is a very good candidate for quantum computing.¹⁷

Suppose now that we can prepare the $^{15}\text{N-V}^-$ center in such a way that all the spin population is in the ground-state level $m_s = |0\rangle$. We have shown that an optically driven Λ transition will change the nuclear-spin projection because of the spin nonconserving transition. Therefore if we optically pump the ground-state level so that we have a Λ transition of the form $|m_s = 0\rangle \rightarrow |m_s = \pm 1\rangle$, then the ground-state popula-

tion will transfer according to the two possible Λ transitions $|m_s = 0\rangle|\uparrow\rangle \rightarrow |m_s = \pm 1\rangle|\downarrow\rangle$ or $|m_s = 0\rangle|\downarrow\rangle \rightarrow |m_s = \pm 1\rangle|\uparrow\rangle$, with flipped nuclear-spin projection (see Fig. 3). Note that both Λ transitions are completely independent from each other. Then, by means of resonance condition, it is possible to produce an arbitrary superposition of the form $|\Psi_w\rangle = \alpha|m_s = 0\rangle + \beta|m_s = 1\rangle$, which represents the qubit.

In order to read this superposition state the electric field F_y needs to be switched off, leading to $\mathcal{A} = 0$. This means that for $F_y = 0$ and weak strain the two-photon transitions are spin conserving, which allows for the detection e.g. $|\alpha|^2$ through the linear response to a σ^+ -polarized laser beam. Single-qubit operations of the qubit state $|\Psi_w\rangle$ can be performed by means of the spin conserving and spin nonconserving transitions, which give rise to phase and amplitude shifts, respectively.

Although our theoretical results have been applied for the N-V centers containing a ^{15}N atom ($I = 1/2$), our model works analogously for ^{14}N ($I = 1$), where Λ transitions of the form $|m_s = 0\rangle|m_I = \pm 1\rangle \rightarrow |m_s = \pm 1\rangle|m_I = 0\rangle$ are achievable at room temperature. Note that this theoretical result proves that the physical process responsible for the Λ transition is the hyperfine interaction, which allows the spin nonconserving transitions in the N-V center in diamond.

In conclusion, we have demonstrated that there exists a Λ transition in $^{15}\text{N-V}^-$ center in diamond that is mediated by the hyperfine interaction which allows us to write quantum information in this solid-state system. Readout can be performed when the electric field is tuned to spin conserving transitions. Our findings clarify the origin of the two-photon transitions observed in Refs. 6 and 7, which is essential for the development of a scalable quantum network made of $^{15}\text{N-V}^-$ centers in diamond, given the possibility to produce a photonic crystal structure in diamond.^{18,19}

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