Laser and Microwave Excitations of Rabi Oscillations of a Single Nitrogen-Vacancy Electron Spin in Diamond

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A collapse and revival shape of Rabi oscillations of a single Nitrogen-Vacancy (NV) center electron spin has been observed in diamond at room temperature. Because of hyperfine interaction between the host 14N nuclear spin and NV center electron spin, different orientation of the 14N nuclear spin leads to a triplet splitting of the transition between the ground m_s=0 and excited states m_s=1. Microwave can excite the three transitions equally to induce three independent nutations and the shape of Rabi oscillations is a combination of the three nutations. This result provides an innovative view of electron spin oscillations in diamond.

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Manipulating single spins in solids is a promising candidate for basic quantum computing implementation [1–3]. The Nitrogen-Vacancy (NV) center in diamond is unique because its electron spin can be polarized and readout optically. More importantly it exhibits extremely long coherence time under optical excitation at room temperature [4–7]. A quantum logical NOT and a conditional two-qubit gate (CROT) using a nearby 13C spin has been presented [8]. Since the first observation of coherently driven electron spin oscillations (Rabi oscillations) of NV center was reported in 2004 [9], several groups have repeated Rabi oscillations measurement as a basic quantum bit manipulation [10–12]. Collapse and revival shape of the Rabi oscillations has been observed in two different works: one is in type Ib diamond with rich nitrogen impurities environment [11], the other is in type IIa diamond with a nearby 13C nuclear spin [13]. In this letter we report our experiments in type IIa diamond (without nearby 13C and very low nitrogen impurity content). We demonstrate a collapse and revival shape of electron spin Rabi oscillations.

Previous theoretical analysis has studied the strong coupling to a nearby nitrogen spin [4,14] as well as nitrogen impurity spin bath [11]. We study the third regime: only taking hyperfine coupling to the NV center’s host 14N nuclear spin into consideration. This is based on low nitrogen content in type IIa diamond. This treatment yields a good explanation for collapse and revival shape observed.

A NV center comprises a substitutional nitrogen atom instead of a carbon atom and an adjacent lattice vacancy. Experiments are carried out in a type IIa bulk diamond (high temperature and high pressure diamond from Sumitomo Electric Industries) with extremely low nitrogen density (∼ 1 ppm). We use a home-built laser scanning confocal microscope system to locate the single NV centers [Fig. 1(a)]. Second order photon correlation function g^2(τ) of center A indicates that it is a single quantum emitter [Fig. 1(b)]. All our experiments are performed at NV center A.

The electronic paramagnetic ground state of NV center is a spin triplet state (3A, S=1). Fig. 1(c) shows the energy level scheme of NV center. As a result of crystall field [11], there is a zero field splitting D=2.87 GHz between sublevels of m_s=0 and m_s=±1(m_s is the projection of spin operator along the z axis). Due to C5v symmetry, m_s=1 and m_s=−1 levels are degenerated.

Strong optical transition between the ground states and the first excited state (also a spin triplet state 3E) is dipole-allowed and the Zero Phonon Line (ZPL) is exhibited at 637 nm (1.945 eV) [15,16]. However, fluores-
three oscillations mentioned above and indicates that all 

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\[ \text{is shown in Fig. 3}. \] We set our microwave frequency res-

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\[ \text{frequencies 8.4 MHz, 6.2 MHz and (c) 4.2 MHz. In all cases } \]

\[ \text{cillations [Fig. 2, black points and lines] for three Rabi }

\[ \text{signal which takes about 1 min.} \]

\[ \text{of } \]

\[ \text{m} = 0 \text{ sublevel via optical pumping [17–19] and readout }

\[ \text{spin state by spin-selective fluorescence [20]. Spin-}

\[ \text{dependent intersystem crossing to a metastable level (singlet, } \]

\[ ^1\text{A} ) \text{ is assumed to be responsible for these differences [4, 21].} \]

In all our experiments, an external static magnetic field of \( \sim 40 \text{ Gauss is applied to split the levels } m_s = \pm 1 \)

\[ \text{sublevels by } \]

\[ 60 \text{ MHz [detailed electron spin resonance spectrum is shown in Fig. 3]. We set our microwave frequency resonant to } m_s = 1 \text{ level. Fig. 1(d) is the pulse sequence for Rabi oscillations measurement. First, the laser is turned on to pump NV center electron spin into } m_s = 0 \text{ state, then the microwave (MW) pulse of different time duration is used to manipulate the electron spin. The final spin state is read out by a second laser pulse. Every cycle of the pulse sequence is about 1 ms, we repeat the measurement cycle for } 10^5 \text{ times to obtain a smooth oscillation signal which takes about 1 min.} \]

We first show a collapse and revival shape of Rabi oscil-

\[ \text{lizations [Fig. 2, black points and lines] for three Rabi}

\[ \text{frequencies 8.4 MHz, 6.2 MHz and 4.2 MHz. In all cases the cohesive oscillations have an envelope: the signal ini-

\[ \text{tially collapses and then revives with decreased amplitude. Weak MW induced oscillation collapses at } t < 1 \mu s \]

\[ \text{while strong MW leads the to a later collapse time } t \sim 1.5 \]

\[ \mu s. \] Fig. 2(d) shows Fast Fourier Transform (FFT) of the three oscillations mentioned above and indicates that all of the three oscillations contain two different frequency components.

R. Hanson et al. [11] observed a collapse and revival shape of oscillations in type Ib diamond with rich nitrogen impurities and regarded it as a result of effective spin bath. However, we use type IIA diamond which contains much lower nitrogen impurities than type Ib and it offers much cleaner spin environment. M.D. Lukin has reported a similar shape of electron spin oscillations in NV center in type IIA diamond and attributed it to the influence of a nearby \(^{13}\text{C}\) nuclear spin [13]. Our Electron Spin Resonance (ESR) spectrum shows a clean environment around the NV center without nearby nuclear spins [Fig. 3(a) and (b)]. A \( \sim 60 \text{ MHz splitting is induced by Zeeman effect. We do not observe splitting caused by } \]

\[ ^{13}\text{C}. \] The ESR spectrum shows a 2.2 MHz triplet splitting, which is due to the hyperfine interaction of electron spin with host \(^{14}\text{N}\) nuclear spin [22] [Fig. 3(b)].

To gain insight into the dynamics of NV center electron spin in clean environment, we consider the hyperfine interaction with host \(^{14}\text{N}\) nuclear spin. The external magnetic field shifted the two \( m_s = \pm 1 \) transitions by \( \sim 60 \text{ MHz}, \) we disregard \( m_s = -1 \) level and treat NV center as a two-level system. We denote \( m_s = 0 \) as level 1 and \( m_s = 1 \) as level 2. The Hamiltonian regarding this hyperfine interaction can be written as: [11]
\begin{equation}
H = A_0 S_0^z I_0^z + A_1 (S_0^z I_0^z + S_0^y I_0^y) - P_0 (I_0^z)^2
\tag{1}
\end{equation}

where \(A_0 = 2.3\) MHz and \(A_1 = 2.1\) MHz are hyperfine coupling tensor parameters, and \(P_0 = -5.1\) MHz is quadrupole splitting for nucleus having spin \(I \geq 1\). The hyperfine energy level scheme \([22, 23]\) is shown in Fig. 3(c); there are three transitions corresponding to three possible \(z\) projections of the \(^{14}\)N nuclear spin and these transitions are separated by \(\alpha_{N} = 2.2\) MHz in energy. We denote middle of the three transitions as \(m_s\) and denote MW frequency in detuning formula \(\delta_f = f_{MW} - f_{0}^{NV}\).

This 2.2 MHz splitting is essentially quasi-static because of slow nuclear spin relaxation (~1 s) compared with fast measurement in a single pulse sequence. In other words, orientation of the host nitrogen nuclear spin does not change during a single measurement cycle. This hyperfine interaction therefore only gives an additional detuning to the resonant MW frequency between \(m_s = 0\) and \(m_s = 1\). We can still treat NV center as a two-level system by non-resonant excitation (adiabatic approximation).

As we repeat pulse sequence for \(10^5\) cycles (~1 minute), the free evolution of \(^{14}\)N gives rise to three possible \(z\) projections of its nuclear spin \((m_f = +1, 0, -1)\). Physically this nuclear spin flips in space, but a static \(N\) nuclear spin model is applicable. The three orientations of \(^{14}\)N nuclear spin have the same probability \(1/3\). MW pulse can excite all the three transitions and they are equally mixed. We average over all three possible Rabi oscillations (ergodic assumption). The Rabi nutation frequencies will arise when the \(^{14}\)N spin is in its three possible projections along the NV axis. As nutations with different frequencies superpose, a beat is present.

Rabi signal considering dephasing process (provided rotating wave approximation is applicable) is an exponent-decayed population change between level 1 and 2 \([24]\):

\begin{equation}
|a_1(t)|^2 = e^{-t/t_0} \frac{f_0^2}{f_e^2} \cos 2\pi f_e t
\tag{2}
\end{equation}

where \(|a_1(t)|^2\) corresponds to population of level 1. \(t_0\) represents decay constant. \(f_0 = \gamma H_R/2\pi\) is the resonant Rabi frequency with \(\gamma\) gyromagnetic ratio of electron and \(H_R\) provided by MW field. \(f_e = (f_0^2 + \delta_f^2)^{1/2}\) is the effective Rabi frequency for a microwave detuning \(\delta_f\). There exist three transitions and three detunings, respectively. Average of the three nutations is:

\begin{equation}
|a_1(t)|^2 = \frac{1}{3} e^{-t/t_0} \sum_{f_e} \frac{f_0^2}{f_e^2} \cos 2\pi f_e t
\tag{3}
\end{equation}

In order to compare this model with experiment, we perform Fast Fourier Transform (FFT) to obtain frequency information [Fig. 2(d)]. We fit the theoretical model (Eq. 3) to experiment curves [Fig. 2, red line].

For short measurement time and large Rabi resonant frequency, it is possible to observe oscillations with lasting decay trend because the first part of a wave packet is just collapse of the nutation. F. Jelezko et al. \([11]\) has reported oscillations with lasting decay tendency. Their Rabi frequency is very high (16 MHz and 39 MHz) and their measurement lasts about \(2 \mu s\). In contrast with their experiments, we do not use very strong MW power to excite nutations with Rabi frequency larger than \(10\) MHz and set our maximum MW duration relatively long (~3.5 \(\mu s\)). Collapse and revival shape should be a general form of Rabi nutations since MW can excite all three transitions within the hyperfine structure as long as the resonant Rabi frequency is bigger than the hyperfine splitting.

We can estimate the characteristic free precession time of \(^{14}\)N. The free precession characteristic time of should be much longer than ~1 ms (the time for a single pulse sequence) and should be much shorter than ~1 min (the time to repeat \(10^5\) cycles). This is a rough estimation and more accurate estimation may be obtained by reducing the cycle number to determine when ergodic assumption will not be applicable.

We also performed spin echo [Fig. 3(d)] and Ramsey fringes [Fig. 3(e)] experiment to determine the spin coherence property of the NV center. The Ramsey fringes are obtained using a microwave pulse sequence \(\pi/2 - t - \pi/2\) for a microwave detuning \(\delta_f = -3.3\) MHz. Our theoretical simulation indicates that the spin dephasing time \(T_2^*\) is about 2.3 \(\mu s\). To eliminate frequency shifts caused by slowly changing nuclear spin environment, we utilize \(\pi/2 - \tau - \pi - \tau - \pi/2\) known as spin echo technique. Fit to the envelop of the spin echo signal (peaks of the curve \(\tau = \tau'\)) gives the coherence time \(\tau_c \approx 4\mu s\). This coherence time for 40 Gauss external static magnetic field is comparable with previous reported values \([10]\).

We demonstrate the response of Rabi oscillations to different MW frequencies [see Fig. 4]. We set the MW detunings to \(\delta_f = 0, 1.1, 2.2, 3.3\) MHz. Hyperfine structure plays an important role here as in the case \(\delta_f = 0\) [see Fig. 2]. Frequency information from FFT of all curves (figures not shown) is in agreement with the effective Rabi frequency \(f_e = (f_0^2 + \delta_f^2)^{1/2}\). The resonant Rabi frequency \(f_0\) of different curves are slightly different (error less than 5%) and may be attributed to the MW power fluctuation. A fit based on equal-weight combination theoretical model is shown in Fig. 4 (red lines). These results demonstrate that the MW frequency detuning can have an influence on determining Rabi period: complicated curve shape may occur under certain detunings. The resonant Rabi frequency and \(\pi\) pulse can
FIG. 4: (color online) Influence of microwave detuning on Rabi oscillations. Rabi oscillations for detuning $\delta_f=0, 1.1, 2.2, 3.3$ MHz. All experiments are carried out under the same microwave power. Fit to exponent decayed sum of three nutations is shown by red line.

be determined correctly only if the hyperfine interaction induced by the host $^{14}$N nuclear spin is considered.

In summary, we have demonstrated the influence of nuclear spin induced hyperfine interaction on Rabi oscillations. Different orientation of the $^{14}$N nuclear spin leads to a triplet splitting of the transition between the ground $m_s=0$ and excited $m_s=1$ states. Microwave can excite the three transitions equally to induce three independent nutations and Rabi signal is a combination of the three nutations. The hyperfine structure due to $^{14}$N exists whatever the NV center’s environment is.

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[1] B. E. Kane, Nature (London) 393, 133 (1998).
[2] R.G. Clark et al., Philos. Trans. R. Soc. London A 361, 1451 (2003).
[3] A.M. Stoneham, A. J. Fisher, and P.T. Greenland, J. Phys. Condens. Matter 15, L447 (2003).
[4] D.A. Redman et al., Phys. Rev. Lett. 67, 3420 (1991).
[5] A. Gruber et al., Science 276, 2012 (1997).
[6] R. Hanson et al., Phys. Rev. Lett. 97, 087601 (2006).
[7] T.A. Kennedy et al., Appl. Phys. Lett. 83, 4190 (2003).
[8] F. Jelezko et al., Phys. Rev. Lett. 93, 130501 (2004).
[9] F. Jelezko et al., Phys. Rev. Lett. 92, 076401 (2004).
[10] L. Childress et al., Science 314, 281 (2006).
[11] R. Hanson et al., Science 320, 352 (2008).
[12] F. Shi et al., arXiv:1002.2465v1 (unpublished).
[13] L. Childress, Ph.D. dissertation. Harvard University, 2007.
[14] T. Gaebel et al., Nat. Phys. 2, 408 (2006).
[15] Y. Mita, Phys. Rev. B 53, 11360 (1996).
[16] J.P. Goss et al., Phys. Rev. B 56, 16031 (1997).
[17] A. P. Nizovtsev et al., Opt. Spectrosc. (USSR) 94, 848 (2003).
[18] T. Gaebel et al., Physica B (Amsterdam) 340, 106 (2003).
[19] J. Harrison, M. J. Sellers, and N. B. Manson, J. Lumin. 107, 245 (2004).
[20] F. Jelezko et al., Appl. Phys. Lett. 81, 2160 (2002).
[21] A. Drabenstedt et al., Phys. Rev. B 60, 11503 (1999).
[22] J. Meijer et al., Appl. Phys. Lett. 87, 261909 (2005).
[23] J. R. Rabeau et al., Appl. Phys. Lett. 88, 023113 (2006).
[24] Elements of Quantum Optics, edited by Pierre Meystre and Murray Sargent (Springer-Verlag, Berlin, 1999).