Nuclear spin induced collapse and revival shape of Rabi oscillations of a single electron spin in diamond*

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A collapse and revival shape of Rabi oscillations in an electron spin of a single nitrogen-vacancy centre has been observed in diamond at room temperature. Because of hyperfine interaction between the host ^{14}N nuclear spin and the nitrogen-vacancy centre electron spin, different orientations of the ^{14}N nuclear spins lead to a triplet splitting of the transition between ground state ($m_s = 0$) and excited state ($m_s = 1$). The manipulation of the single electron spin of nitrogen-vacancy centre is achieved by using a combination of selective microwave excitation and optical pumping at 532 nm. Microwaves can excite three transitions equally to induce three independent nutations and the shape of Rabi oscillations is a combination of the three nutations.

Keywords: nitrogen-vacancy centre, single electron spin, Rabi oscillations, hyperfine interaction

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1. Introduction

Manipulating single spins in solid is a promising candidate for basic quantum computing implementation.[1−3] The nitrogen-vacancy (NV) centre 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 ^{13}C spin have been presented.[8] Since the first observation of coherently driven electron spin oscillations (Rabi oscillations) of NV centre was reported in 2004,[9] several groups have repeated Rabi oscillation measurement as a basic quantum bit manipulation.[10−12] Collapses and revival shapes of the Rabi oscillations have been observed in two types of samples: Ib diamond being rich in single substitutional nitrogen atoms[11] and nitrogen-free IIa diamond with a nearby ^{13}C nuclear spin.[13] In addition, state collapses and revivals in a superconducting charge qubit and in the entanglement of two qubits coupled to a thermal reservoir have been investigated.[14,15] In this paper, we report on our experiments in type IIa diamond (without nearby ^{13}C and very low nitrogen impurity content). We demonstrate a collapse and revival shape of electron spin Rabi oscillations. The shape of Rabi oscillations is a combination of three nutations. This result provides an innovative view of electron spin oscillations in diamond.

The strong coupling to a nearby nitrogen spin[6,16] as well as nitrogen impurity spin bath has been studied theoretically before.[11] We study a third regime, where only hyperfine coupling to the host ^{14}N nuclear spin of NV centre is taken into consideration. This is based on low nitrogen content in type IIa diamond. This treatment yields a good explanation for the collapse and revival shape observed.

2. Experiment

An NV centre comprises a substitutional nitrogen atom instead of a carbon atom and an adjacent lattice vacancy. There are two charged states of the NV centre, namely NV^0 with zero net charge and NV^- with one additional electron. We conduct our experiments on NV^-. Experiments are carried out in a type-IIa bulk diamond (high temperature and high pressure diamond from Sumitomo Electric Industries) with low nitrogen density ($\sim$ 1 ppm). We use a home-made

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laser scanning confocal microscope system to locate the single NV centres (Fig. 1(a)). A linearly polarized 532 nm continuous-wave laser modulated by an acoustic optical modulator (AOM) with a rising edge of 10 ns is focused onto the sample along the [001] crystal axis with an oil immersion objective lens (numerical aperture=1.3). The fluorescence is collected by the same objective lens and passes through a 650 nm long-pass filter. Signal is detected by a single photon counting module (SPCM, Perkin-Elmer) associated with a National Instruments counter card 6602. The operation mode of the scanning confocal microscope system is to scan one dimension while the other dimension is fixed. Second order photon correlation function $g^2(\tau)$ of centre A indicates that it is a single quantum emitter (Fig. 1(b)). All our experiments are performed at NV centre A.

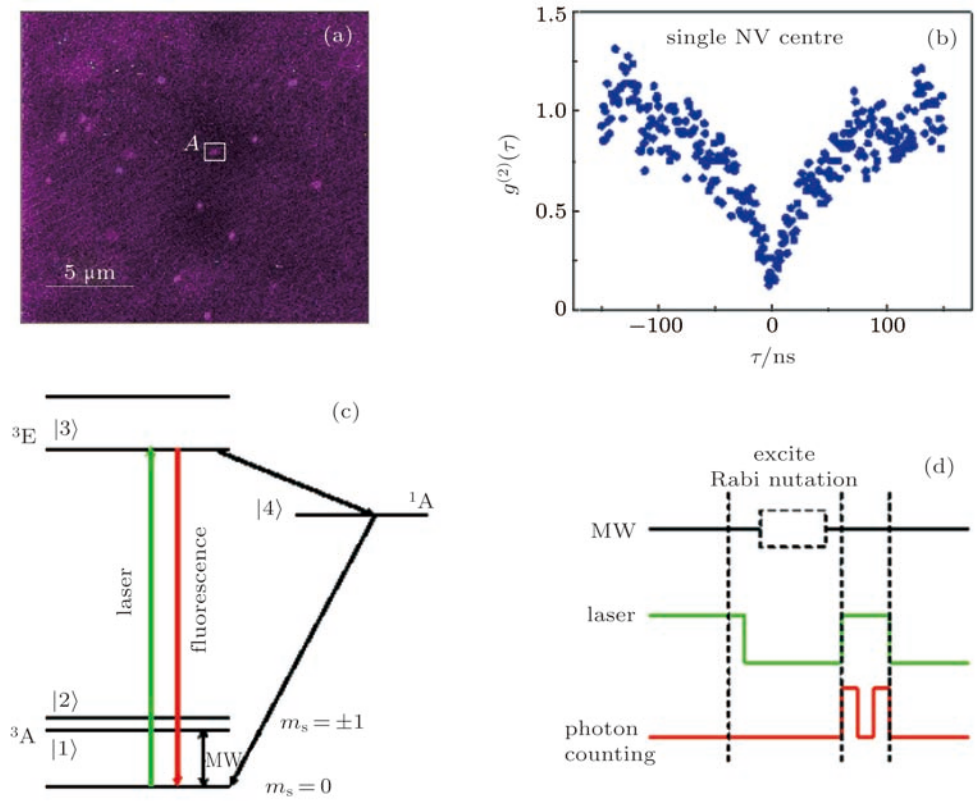


Fig. 1. (colour online) Optical properties of NV centre and Rabi oscillations pulse sequence. (a) A scanning confocal image of the sample. Bright spot A corresponds to the NV centre we investigate (in all our experiments). (b) The second order photon correlation function $g^2(\tau)$. (c) The energy level scheme of the NV centre in diamond. (d) The pulse sequence for Rabi oscillation experiment. Optical pumping lasts for about 3 μs, the NV centre is polarized into $m_s = 0$ sublevel. Microwave manipulation is used to drive Rabi oscillations. The minimum duration of MW pulse is 25 ns and the maximum duration is about 3.5 μs.

The electronic paramagnetic ground state of NV centre is a spin triplet state (^{3}A, S= 1). Figure 1(c) shows the energy level scheme of NV centre. As a result of crystal 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 C_{3V} symmetry, $m_s = 1$ and $m_s = -1$ levels are degenerate.

Strong optical transition between the ground states and the first excited state (also a spin triplet state ^{3}E) is dipole-allowed and the zero phonon line (ZPL) is exhibited at 637 nm (1.945 eV).[17,18] However, the fluorescence intensity of a single NV centre is strongly dependent on its spin state. The average photon emission rate is quite smaller for transitions involving the $m_s = \pm 1$ levels than for the $m_s = 0$ level, which allows polarization to $m_s = 0$ sublevel via optical pumping[19−21] and readout of the spin state by spin-selective fluorescence.[22,23] Spin-dependent intersystem crossing to a metastable level (singlet, ^{1}A) is assumed to be responsible for these differences.[4,24]

In all our experiments, an external static magnetic field of ∼ 40 Gauss (1 Gauss=10^{-4} T) is applied to split $m_s = \pm 1$ sublevels by ∼ 60 MHz. We set our

microwave frequency resonant to $m_s = 1$ level. Figure 1(d) shows the pulse sequence for Rabi oscillations in measurement. First, the laser is turned on to pump the electron spin of the NV centre into $m_s = 0$ state, then the microwave (MW) pulses of different durations are used to manipulate the electron spin. The final spin state is read out by a second laser pulse. Each cycle of the pulse sequence is about 1 ms. We repeat the measurement cycle 10^5 times, which takes about 1 min, to obtain a smooth oscillation signal.

3. Results and discussion

We first show a set of Rabi oscillations with collapses and revival shape (Fig. 2, black points) at three Rabi frequencies 8.4 MHz, 6.2 MHz and 4.2 MHz. In all the cases the coherent oscillations each have an envelope: the signal initially collapses and then revives with decreased amplitude. Weak MW induced oscillation collapses at $t < 1$ μs while strong MW leads to a later collapse time $t \sim 1.5$ μs. Figure 2(d) shows fast Fourier transform (FFT) of the three oscillations mentioned above and indicates that the oscillations in Figs. 2(a) and 2(c) contain two different frequency components. The singlet of FFT (Fig. 2(b)) is due to imperfect fitting. Our FFT resolution is limited by the total measurement time. Figure 2 shows three Rabi nutations with time up to 3.5 μs. This time gives an FFT spectrum resolution of around 0.3 MHz.

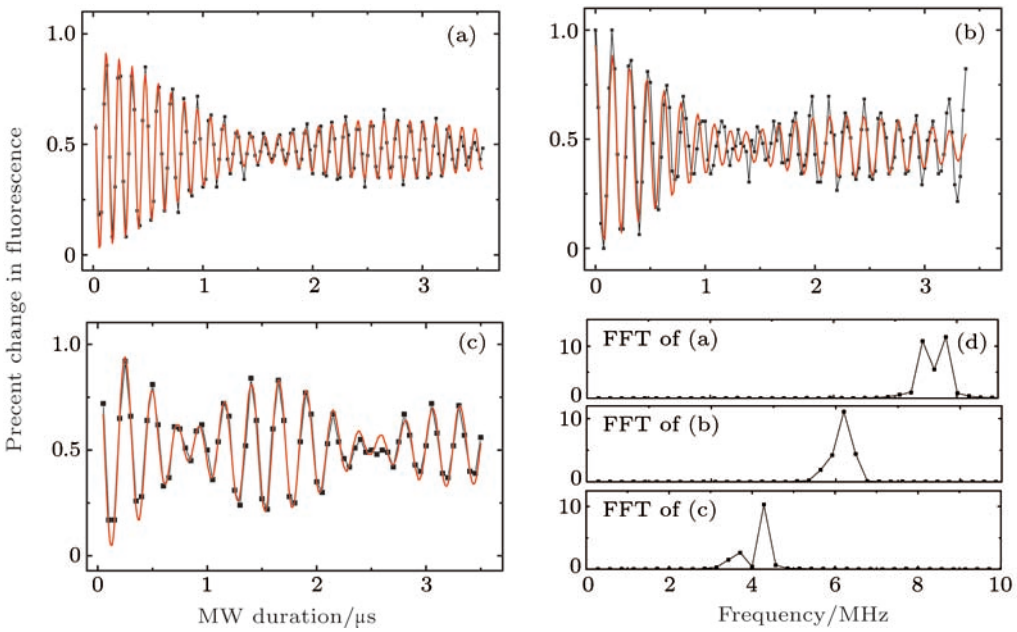


Fig. 2. (colour online) Collapses and revival shapes of Rabi oscillations. Experimental data are denoted as solid dots. In calculations considered are the couplings to host ^{14}N nuclear spin for Rabi frequencies of (a) 8.4 MHz, (b) 6.2 MHz and (c) 4.2 MHz. Red lines represent the fittings to an exponentially decayed sum of three cosines with different frequencies (see Eq. (3) in text). All curves are obtained at MW detuning $\delta_f = 0$. The laser power is 200–300 μW for all curves. Panel (d) shows the fast Fourier transform of results in panels (a), (b) and (c).

Hanson *et al.*[11] observed that oscillations with a collapse and revival shape in type-Ib diamond (with rich nitrogen impurity content). From the theoretical analysis they found that the nitrogen nuclear spin of the NV centre plays an essential role in the observed dynamics. However, we use type-IIa diamond which contains much lower nitrogen impurity content than type-Ib and it offers much cleaner spin environment. Childress[13] has reported on a similar shape of electron spin oscillations in NV centre in type-IIa diamond and attributed it to the influence of a nearby ^{13}C nuclear spin. Our optically detected magnetic resonance (ODMR) spectrum shows a clean environment around the NV centre without nearby nuclear spins (Figs. 3(a) and 3(b)). A $\sim$ 60 MHz splitting is induced by Zeeman effect. We have not observed the splitting caused by ^{13}C. The ODMR spectrum shows a 2.2 MHz triplet splitting, which is due to the hyperfine interaction of electron spin with host ^{14}N nuclear spin[25] (Fig. 3(b)).

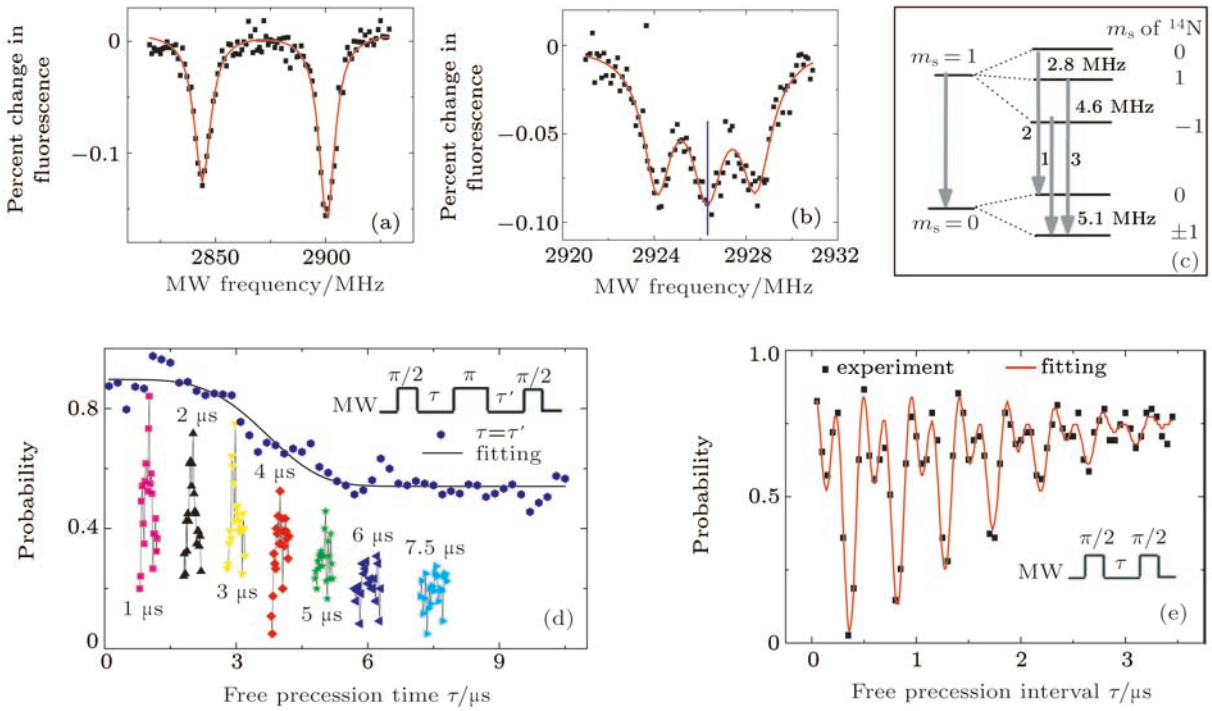


Fig. 3. (colour online) ODMR spectrum, hyperfine energy level, Ramsey fringes and spin echo. (a) ODMR spectrum of the NV centre. Two main peaks correspond to $m_s = 1$ and $m_s = -1$ respectively. (b) ODMR under weak MW and laser power, triplet splitting of 2.2 MHz, which is an indication of the host ^{14}N. Red curves represent the fitting in the form of sum of three Lorentz functions. (c) Theoretically calculated hyperfine energy level scheme. The three allowed hyperfine transitions are numbered and correspond to three ODMR signal troughs in panel (b). (d) Spin echo signal in a low magnetic field ($\sim 0.4 \times 10^{-2}$ T). (e) Ramsey fringes signal. MW pulse sequences in panels (d) and (e) are depicted as an inset, and the resonant Rabi frequency for spin echo experiment and that for Ramsey fringes experiment are both about 4 MHz.

To gain an insight into the electron spin dynamics of NV centre in a clean environment, we consider the hyperfine interaction of electron spin with host ^{14}N nuclear spin. The external magnetic field shifts the two $m_s = \pm 1$ transitions by ~ 60 MHz, we disregard $m_s = -1$ level and treat NV centre 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]

$$H = A_0 S_0^z I_0^z + A_1 (S_0^x I_0^x + S_0^y I_0^y) - P_0 (I_0^z)^2, \quad (1)$$

where A_0 and A_1 are hyperfine coupling tensor parameters, and P_0 is quadrupole splitting for nucleus having spin $I \geq 1$. In this case, nitrogen nucleus has $A_0 = 2.3$ MHz, $A_1 = 2.1$ MHz and $P_0 = -5.1$ MHz. The hyperfine energy level scheme[25,26] is shown in Fig. 3(c), where there are three transitions corresponding to three possible z projections of the ^{14}N nuclear spin and these transitions are separated by $\alpha_N \approx 2.2$ MHz in energy. We denote the frequency of middle transition of the three transitions as $\omega_0 = 2\pi f_0^{\rm NV}$ and denote MW frequency in detuning formula $\delta_f = f_{\rm MW} - f_0^{\rm NV}$.

This 2.2 MHz splitting is essentially quasi-static because of slow nuclear spin relaxation ($\sim$ 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 gives only an additional detuning to the resonant MW frequency between $m_s = 0$ and $m_s = 1$. We can still treat NV centre as a two-level system under non-resonant electromagnetic excitation (adiabatic approximation).

As we repeat the pulse sequence for 10^5 cycles (~ 1 min), the free evolution of ^{14}N gives rise to three possible z projections of its nuclear spin ($m_I = +1$, 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 take an

average over all three possible Rabi oscillations (ergodic assumption). The Rabi nutation frequency will arise when the ^{14}N spin is in its three possible projections along the NV axis. As nutations with different frequencies are superposed, a beat frequency is present.

Dephasing of the Rabi signal follows the exponential relaxation of the population difference (provided rotating wave approximation is applicable) between levels 1 and 2.[27] The following expression

$$|a_1(t)|^2 = \mathrm{e}^{-t/t_0}\frac{f_0^2}{f_\mathrm{e}^2}\cos 2\pi f_\mathrm{e} t \tag{2}$$

corresponds to population of level 1, where t_0 represents decay constant, $f_0 = \gamma H_\mathrm{R}/2\pi$ is the resonant Rabi frequency with γ being the gyromagnetic ratio of electron and H_R denoting the magnetic field provided by MW field, $f_\mathrm{e} = (f_0^2+\delta_f^2)^{1/2}$ is the effective Rabi frequency for a microwave detuning δ_f. There exist three transitions and three detunings, respectively. The average value of the three nutations is

$$|a_1(t)|^2 = \frac{1}{3}\mathrm{e}^{-t/t_0}\sum_{f_\mathrm{e}}\frac{f_0^2}{f_\mathrm{e}^2}\cos 2\pi f_\mathrm{e} t. \tag{3}$$

In order to compare the result from this model with experimental result, we perform FFT to obtain frequency information (Fig. 2(d)). We fit the theoretical model ((Eq. (3)) to experimental curves (red line in Fig. 2).

For short measurement time and large Rabi resonant frequency, it is possible to observe oscillations with a lasting decay trend because the first part of a wave packet is just collapse of the nutation. Jelezko *et al.*[9] reported on the oscillations with lasting decay tendency. Their Rabi frequencies were very high (16 MHz and 39 MHz) and their measurement lasted about 2 μs. In contrast with their experiments, we do not excite nutations with Rabi frequency larger than 10 MHz and set our maximum MW duration relatively long ($\sim$ 3.5 μs). The collapse and revival shape should be in 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 higher than the corresponding frequency of hyperfine splitting.

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

We also perform spin echo (Fig. 3(d)) and Ramsey fringes (Fig. 3(e)) experiment to determine the spin coherence property of the NV centre. The Ramsey fringes are obtained using a microwave pulse sequence $\pi/2$–τ–$\pi/2$ for a microwave detuning $\delta_f = -3.3$ MHz. Details of the Ramsey technique can be found in Ref. [28]. For a two-level system, the Ramsey sequence can cause a population oscillation with a frequency equal to the microwave detuning δ_f. Our theoretical simulation indicates that the spin dephasing time T_2^* is about 2.3 μs. To eliminate frequency shifts caused by slowly changing nuclear spin environment, we utilize $\pi/2$–τ–π–τ'–$\pi/2$, known as the spin echo technique.[29] Extra τ' appears in the spin echo sequence. If the two wait parameters are equal, $\tau = \tau'$, the environmental random phase shift factors out. Fitting to the envelop of the spin echo signal (peaks of the curve $\tau = \tau'$) gives the coherence time $\tau_c \approx 4$ μs. This coherence time for 0.4×10^{-2} T 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 are also aware that the influence of detuning in a linear ion trap has been studied.[30] Here, 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 $f_\mathrm{e} = (f_0^2 + \delta_f^2)^{1/2}$. 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 π pulse can be determined correctly only if the hyperfine interaction induced by the host ^{14}N nuclear spin is considered.

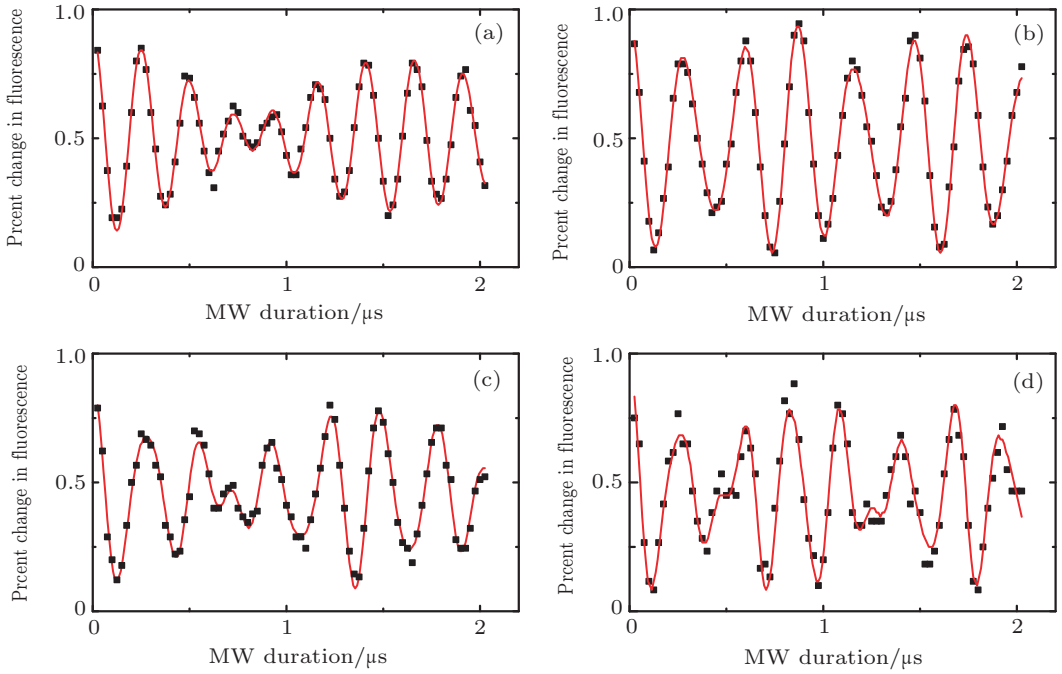


Fig. 4. (colour online) Influence of microwave detuning on Rabi oscillations. Rabi oscillations for detuning δ_f of 0 (a), 1.1 MHz (b), 2.2 MHz (c), 3.3 MHz (d). All experiments are carried out under the same microwave power. Fitting to a superposition of three nutations with exponential-decay tendency is shown by red line.

4. Conclusion

We demonstrated the influence of nuclear spin induced hyperfine interaction on Rabi oscillations. The manipulation of single electron spin of the NV centre is achieved by using a combination of selective microwave excitation and optical pumping at 532 nm. Different orientations of the ^{14}N nuclear spins lead to a triplet splitting of the transition between ground state ($m_s = 0$) and excited state ($m_s = 1$). Microwave can excite 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, no matter what environment the NV centre is in. In previous relevant work[11,13,31] the use of type-Ib diamond makes it harder, because the effects caused by spin bath or the host ^{14}N need to be separated from each other through a cleaner and purer environment. In our experiment, type-IIa diamond is used and the environment around the NV centre we investigate is very clean. This can ensure that what we observe in experiment can be induced only by internal ^{14}N nuclear spin.

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