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Measurement of transverse hyperfine interaction by forbidden transitions

Mo Chen (陈), 1,2 Masashi Hirose,1,3 and Paola Cappellaro1,3,*

1Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
2Department of Mechanical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA
3Department of Nuclear Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA

(Received 14 April 2015; revised manuscript received 21 May 2015; published 6 July 2015)

Precise characterization of a system’s Hamiltonian is crucial to its high-fidelity control that would enable many quantum technologies, ranging from quantum computation to communication and sensing. In particular, nonsecular parts of the Hamiltonian are usually more difficult to characterize, even if they can give rise to subtle but non-negligible effects. Here we present a strategy for the precise estimation of the transverse hyperfine coupling between an electronic and a nuclear spin, exploiting effects due to nominally forbidden transitions during the Rabi nutation of the nuclear spin. We applied the method to precisely determine the transverse coupling between a nitrogen-vacancy center electronic spin and its nitrogen nuclear spin. In addition, we show how this transverse hyperfine coupling, which has been often neglected in experiments, is crucial to achieving large enhancements of the nuclear Rabi nutation rate.

DOI: 10.1103/PhysRevB.92.020101 PACS number(s): 76.70.Hb, 03.67.Lx, 76.60.—k, 76.70.Dx

Quantum technologies promise to revolutionize many fields, ranging from precision sensing to fast computation. The success of novel technologies based on quantum effects rests on engineering quantum systems robust to noise and decoherence and on controlling them with high precision. Solid-state systems comprising nuclear spins have emerged as promising candidates, since the nuclear spin qubits are only weakly coupled to external fields and thus exhibit long coherence times. In order for nuclear spins to be used as good qubits, there are two important requirements: Their Hamiltonians need to be known with very high precision, as this would enable applying, e.g., optimal control methods [1,2], and strong driving should be available, in order to achieve fast gates. Here we show how to meet these two requirements by exploiting nominally forbidden transitions in a hybrid electronic-nuclear spin system associated with the nitrogen-vacancy center in diamond [3]. Specifically, we use second-order effects due to mixing of the electronic and nuclear spin states [4] in order to identify with high precision their coupling strength and to enhance the nuclear spin nutation rate [5].

The nitrogen-vacancy (NV) center is a naturally occurring point defect in diamond [6]. Owing to its optical properties and long coherence times, it has emerged as a versatile system given by the electronic spin of the NV center ($S$) and the nuclear spin ($I$) of the nitrogen atom. Precise knowledge of the hyperfine interaction tensor $A$ would enable achieving more precise control, elucidating modulations of the NV echo dynamics or, as we show here, achieving faster Rabi nutation of the nuclear spin.

**Theoretical model.** The NV ground state is a two-spin system given by the electronic spin of the NV center ($S = 1$) and the nuclear spin ($I = 1$) of the substitutional $^{14}$N adjacent to the vacancy that comprise the defect. In the experiments, we are only interested in two of the nuclear spin levels ($m_I = \pm 1.0$) that we drive on resonance, while the third level can be neglected. Then, the Hamiltonian of the reduced system [34,35] is given by $\mathcal{H} = \mathcal{H}_{||} + \mathcal{H}_{\perp}$, where the secular, $\mathcal{H}_{||}$, and nonsecular, $\mathcal{H}_{\perp}$, terms are

$$\mathcal{H}_{||} = \Delta S_z^2 + \left( \gamma_e B_z + \frac{A_z}{2} \right) S_z + \left( Q + \gamma_n B_z \right) I_z + A_I S_z I_z,$$

$$\mathcal{H}_{\perp} = \sqrt{2} A_{\perp} (S_z I_x + S_y I_x).$$

Here, $S$ and $I$ are the electron spin-1 and nuclear spin-1/2 operator, respectively. Also, $\Delta = 2.87$ GHz is the zero-field splitting and $Q = -4.945$ MHz [23] is the nuclear quadrupolar interaction. The NV spin is coupled to the nuclear spin by a hyperfine interaction with a longitudinal component $A_z = -2.162$ MHz [23] and a transverse component $A_{\perp}$ which we...
want to estimate. A magnetic field $B_z$ is applied along the NV crystal axis [111] to lift the degeneracy of the $m_z = \pm 1$ level, yielding the electron and nuclear Zeeman frequencies $\gamma_eB_z$ and $\gamma_nB_z$, where $\gamma_e = 2.8$ MHz/G and $\gamma_n = -0.308$ kHz/G. Let $|m_e,m_n\rangle$ be eigenstates of $\hat{H}_\text{e}$. The transverse coupling $A_{\perp}$ mixes states connected via zero-quantum (ZQ) transitions, $|+1,0\rangle \leftrightarrow |0,1\rangle$ and $|0,0\rangle \leftrightarrow |-1,1\rangle$. Diagonalization of the total Hamiltonian can then be achieved by rotating the two ZQ subspaces with a unitary transformation $U_{\text{ZQ}} = e^{-i(\sigma^+_{\perp}t + \gamma_nB_z t)}$, where we defined $\sigma^+_{\perp} = i(|+1,0\rangle\langle0,1| + |0,1\rangle\langle1,0|), \sigma^-_{\perp} = i(|0,0\rangle\langle-1,1| + |-1,1\rangle\langle0,0|)$, and the rotation angles are

$$\tan(2\theta^+) = \frac{2A_{\perp}}{\Delta + \gamma_eB_z - \gamma_nB_z - Q},$$
$$\tan(2\theta^-) = \frac{-2A_{\perp}}{\Delta - \gamma_eB_z - A_{||} + \gamma_nB_z + Q}. \quad (2)$$

Because of this level mixing, a field on resonance with the nuclear spin transition also drives electronic transitions. Although the electronic spin state is unchanged to first order, the nuclear spin transition also drives electronic transitions. As a result, a 1 $\mu$s laser excitation polarizes the NV-14N system into the $|0,1\rangle$ state.

Then, the NV spin is prepared in the desired Zeeman state by a strong microwave (MW) pulse ($\tau_p \approx 50$ ns) before coherently driving the nuclear spin by an rf field on resonance with the nuclear transition $|m_e,1\rangle \leftrightarrow |m_e,0\rangle$, for a duration $\tau$ (see Fig. 1). Finally, the nuclear spin state is detected by employing a MW selective pulse ($\tau_p \approx 700$ ns) that maps the nuclear spin state onto the NV spin, which in turn can be read out optically due to spin-dependent fluorescence emission intensity. The nuclear Rabi oscillations in Fig. 2 clearly show that for a fixed driving strength, the effective Rabi frequency is quite different in the three electronic spin manifolds.

Here, $\alpha_m$ denote the enhancement factors in each manifold of the NV spin,

$$\alpha_{+1} \approx 1 + \frac{\gamma_e}{\gamma_n} \frac{A_{\perp}}{\Delta + \gamma_eB_z - \gamma_nB_z - Q},$$
$$\alpha_0 \approx 1 + \frac{\gamma_e}{\gamma_n} \frac{A_{\perp}}{\Delta + \gamma_eB_z - \gamma_nB_z - Q} + \frac{A_{\perp}}{\Delta - \gamma_eB_z - A_{||} + \gamma_nB_z + Q},$$
$$\alpha_{-1} \approx 1 + \frac{\gamma_e}{\gamma_n} \frac{A_{\perp}}{\Delta - \gamma_eB_z - A_{||} + \gamma_nB_z + Q}, \quad (7)$$

where we show expressions exact up to the first order in $\phi^\pm$ (see Ref. [34] for the exact expressions). The Hamiltonian $\hat{H}_e$ can be neglected since electronic spin transitions are far off resonance.

Owing to the strong dependence of the enhancement factors on the transverse hyperfine coupling, we can determine $A_{\perp}$ with high precision from measurement of the 14N Rabi oscillations.

**Experiments.** We used a home-built confocal microscope to measure the transverse hyperfine interaction of a single NV center in an electronic grade diamond sample (Element Six, 14N concentration $n_{14N} < 5$ ppb, natural abundance of 13C). The NV center is chosen to be free from close-by 13C. We worked at magnetic fields (300–500 G) close to the excited state level anticrossing so that during optical illumination at 532 nm, polarization of the NV spin can be transferred to the nuclear spin by their strong hyperfine coupling in the excited state [36]. As a result, a 1 $\mu$s laser excitation polarizes the NV-14N system into the $|0,1\rangle$ state.
To confirm the expected dependence of the Rabi enhancement factors on the external magnetic field and the NV state, we measured the Rabi oscillations at the three electronic spin manifolds with varying magnetic field $B_x$. As shown in Fig. 3, the measured Rabi frequencies match well with the theoretical model. It is worth noting that contrary to the static pseudonuclear Zeeman effect [4], there is a large enhancement ($\alpha_0 \approx 16$, $\alpha_{\perp} \approx -9$) even at zero field. Also, close to the ground state avoided crossing ($B \approx 0.1 \, \text{T}$), the enhancement can become very large, exceeding 100. The validity of our approximation in this regime can be confirmed by numerical simulations [34].

While these experiments could be used to extract $A_\perp$, this is not a practical method to obtain a good enough estimate. The range of magnetic field is restricted by the need to be close to the excited state level anticrossing to achieve a good polarization of the nuclear spin. The number of acquired points is limited by the time it takes to change and properly align the external magnetic field. In addition, there might be variations in the bare Rabi frequency in the three manifolds, because of different responses of the electronics used to drive the nuclear spins at the different frequencies.

In order to avoid these difficulties, we fixed the magnetic field to 509 G and instead linearly swept the amplitude of the rf driving ($B_1$). With this procedure, we do not need an independent measure of the bare Rabi frequency in order to extract the transverse hyperfine coupling strength. The relative rf amplitudes $B_1$ obtained when varying the driving strength can be measured at each nuclear resonance frequency by monitoring the rf voltage with an oscilloscope, confirming its linear dependence with applied power.

We thus measure the effective nuclear Rabi frequency as a function of the normalized rf amplitude $B_1 / \left| B_{1,\text{max}} \right|$ in all three electronic manifolds (Fig. 4). The measured Rabi frequency $\Omega_m$ is related to its on-resonance value by $\Omega_m = \sqrt{\Omega^2 + \delta^2}$, where $\delta$ is the detuning from the nuclear spin resonance frequency. We incorporate this unknown, small detuning in our model and fit the experimental data with the Rabi enhancement formulas (5)–(7). From the fit, we obtain an estimate of the transverse hyperfine coupling, $A_\perp = -2.62 \pm 0.05 \, \text{MHz}$, in good agreement with recently published values and with better precision.

In order to achieve even better precision, we need to consider all the sources of uncertainty and errors. We find that small errors from imperfect MW $\pi$ pulses and nuclear polarization only contribute to a reduced fluorescent contrast, but do not affect the estimate of the Rabi frequency under our experimental condition. The detuning of the selective MW and rf pulses from resonance and uncertainty in $A_\parallel$ contributes only linearly to the uncertainty. All these minor errors and uncertainties hardly affect the final uncertainty in the estimate of $A_\perp$ [34]. The major source of error arises instead from the uncertainty in the measured Rabi frequency, which is limited by the photon shot noise of the optical readout process. Therefore, the precision of the estimate could be improved with more averaging, at the expense of a longer measurement time. Currently, our total measurement time is limited by the stability of the experimental setup, yielding $\delta A_\perp \sim 50 \, \text{kHz}$. Improving the stability of the setup by reducing thermal fluc-
tations and noise in the driving field (also using decoupling schemes [37,38]), or by employing small ensembles or more efficient optical readout methods such as solid-immersion lenses [39] and charge-state sensing [40], could provide higher precision. Then, the limit would come from uncertainties in $\gamma_e$ and $\gamma_n$, with a relative error of $10^{-4}$ [26,41], yielding an uncertainty in $A_\perp$ of a few hundred Hz [34].

Conclusions. In conclusion, we observed enhanced nuclear Rabi oscillations in the NV-14N system due to level mixing between electronic and nuclear spin states. We harness the strong dependence of this enhancement on the transverse hyperfine coupling to determine its value with high precision. Theoretical analysis predicts an enhancement factor of almost three orders of magnitude when the magnetic field is close to the ground state level anticrossing, promising fast manipulation of the nuclear spin qubit at ~MHz rates, with only moderate driving strengths. More broadly, the method presented here can be applied to many other electron-nuclear hybrid spin systems to similarly characterize their interaction Hamiltonian with high precision. Our results indicate that taking into account the nonsecular parts of a system’s Hamiltonian is crucial to achieving faster and more accurate control of the quantum system.

This work was supported in part by the U.S. Air Force Office of Scientific Research Grant No. FA9550-12-1-0292 and by the U.S. Office for Naval Research Grant No. N00014-14-1-0804. M.C. and M.H. contributed equally to this work.


