

# Extending and measuring dephasing times of nuclear spins in NV centers of diamond

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Long coherence times rank among the most important performance measures for many different types of quantum technology. In the electron-nuclear spin system of NV centers of diamond, the nuclear spins provide particularly long dephasing times. However, since initialization and readout require assistance from the electron spin, the apparent dephasing times can be reduced by the electron spin lifetime. Here we propose and implement schemes for measuring and extending the dephasing times of nuclear spins, resulting in dephasing times that are longer than the longitudinal relaxation time  $T_1$  of the electron spin.

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## I. INTRODUCTION

In all areas of quantum technologies like quantum computing and quantum sensing [1], the preservation of quantum information in the presence of a noisy environment is one of the most challenging tasks. Hybrid systems, such as the combination of electron spin and the nearby nuclear spin of the nitrogen-vacancy (NV) centers in diamond [2,3], are promising for implementing quantum sensors [4–7] and quantum computers [8–10], which are believed to have a large potential for speeding up the solution of certain problems that have no easy solutions for classical computers. One of the main benefits of the NV system lies in the long coherence time of the nuclear spins, e.g., up to one minute at low temperature (3.7 K) [11].

However, the preservation and measurement of the coherence are limited by the longitudinal relaxation time ( $T_1^e$ ) of the electron spin [12–15]. Specifically, if the electron spin changes its state as a result of longitudinal ( $T_1^e$ ) relaxation, the connected nuclear spin experiences a different effective magnetic field due to the hyperfine coupling and therefore a different precession frequency, which results in the loss of coherence [14]. In addition, the detection of the coherence of the nuclear spin depends on the polarization of the electron spin and the loss of this polarization due to longitudinal relaxation reduces the measured coherence time [12,13]. Suitable laser pulses can restore the electron spin polarization. However, this strategy works only in the cases of sufficiently weak hyperfine couplings, where the dephasing and depolarization

of the nuclear spins caused by such additional laser pulses are not significant [15–18]. It has already been demonstrated that nuclear spin coherence can exceed the electron spin lifetime when the coupling is small ( $\sim 2.66$  kHz) [15].

In this article, we propose and implement alternative schemes that help for preserving and measuring the nuclear coherence beyond the electron spin lifetime  $T_1^e$ , in systems whose hyperfine couplings are two orders of magnitude stronger than in previous work [15]. Therefore, our strategy is more suitable in quantum technological applications, where the stronger couplings are crucial for speeding up the control of the nuclear spins. While there are experiments that use  $>100$  kHz coupling strengths at low and room temperatures [13,19,20], those works do not show coherence times longer than  $T_1^e$ . Since the effect of the electron spin relaxation on the nuclear spin coherence increases with the strength of the hyperfine coupling [13,16], the implementation of our strategy is also more challenging. To overcome this challenge, we use efficient quantum control of nuclear spins based on the hyperfine interaction [21–23] in two ways: (i) we refocus the nuclear spin coherence by generating a Hahn echo; (ii) we modify the detection scheme: instead of measuring the coherence of the nuclear spin in only one subspace of the electron state [4,12], we exchange the states of the nuclear and electron spins using a SWAP gate [8], which transfers the nuclear spin coherence into an electronic spin transition. From there, it can be measured through the combination of a  $\pi/2$  microwave (MW) pulse and a readout laser pulse. Moreover, in contrast to earlier work based on encoded qubits [17,24], our current work directly uses the physical qubits, thereby reducing the required resources. While it is possible to work at low temperature where  $T_1^e$  can become much longer [25,26], we choose here to work at room temperature, which is an important prerequisite for many applications, e.g., in quantum sensing.

## II. SYSTEM

The spin system consists of the electron spin, the intrinsic  $^{14}\text{N}$  nuclear, and one  $^{13}\text{C}$  nuclear spin. Our goal is to

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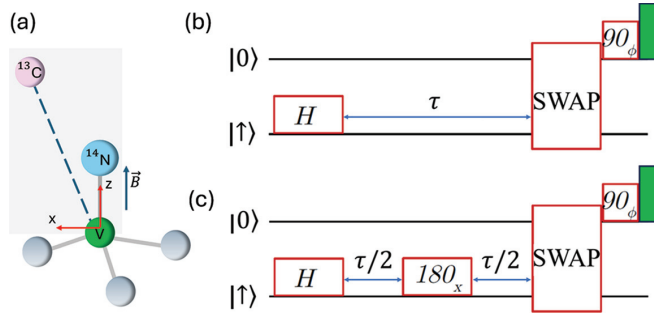


FIG. 1. (a) Geometry and coordinate system for the electron (NV) and  $^{13}\text{C}$  spins, with the NV and  $^{13}\text{C}$  spin in the  $xz$  plane [22,44]. (b),(c) Quantum circuits for measuring the dephasing of the  $^{13}\text{C}$  spin during free precession (b) and with refocusing by a Hahn echo (c). In each circuit, the top line denotes the electron spin qubit and the bottom the  $^{13}\text{C}$  spin. The initial state is  $|0, \uparrow\rangle$ .  $H$  represents a Hadamard gate,  $180_x$  the refocusing operation (echo), and SWAP the two-qubit SWAP gate. The filled rectangles indicate the readout laser pulses.

extend and measure the coherence of the  $^{13}\text{C}$  nuclear spin. Perturbations of the electron and  $^{13}\text{C}$  nuclear spins from the  $^{14}\text{N}$  spin must be minimized. One method is to polarize the  $^{14}\text{N}$  into one state, such as  $m_N = 1$ , using the level anticrossing [27–29]. In our present work with the static magnetic field  $B \approx 148$  G, the  $^{14}\text{N}$  is partially polarized, i.e., the population of the state  $m_N = 1$  is about 60% [see the Supplemental Material (SM) (Sec. II A 2, Fig. 5)] [30]. Therefore, we focus on the subsystem where the  $^{14}\text{N}$  is (and remains) in the  $m_N = 1$  state [21,41,42]. This is a safe assumption since the population lifetime ( $T_1^N$ ) of the  $^{14}\text{N}$  nuclear spin is much longer than that of the electron spin [17,43]. The details on initializing and normalizing this subsystem are presented the SM (Sec. II) [30]. In implementing the control for the electron and  $^{13}\text{C}$  spins, we use the MW pulses with a Rabi frequency of approximately 0.5 MHz, which is small compared to hyperfine coupling between the electron and  $^{14}\text{N}$  spin. Accordingly, they only drive the transitions from the electron states  $m_S = 0$  to one of the  $m_S = \pm 1$  and are selective for  $m_N = 1$  [21,42]. The spins interact with a magnetic field  $B$  oriented along the symmetry axis of the NV center. In a suitable reference frame, shown in Fig. 1(a), we can write the relevant part of the Hamiltonian as

$$\frac{1}{2\pi} \mathcal{H}_{e,C} = DS_z^2 - (v_e - A_N)S_z - v_C I_z + A_{zz}S_z I_z + A_{zx}S_z I_x, \quad (1)$$

where  $S_z$  denotes the electron spin-1 operator and  $I_{x,z}$  the  $^{13}\text{C}$  spin-1/2 operators. The zero-field splitting is  $D = 2.87$  GHz.  $v_{e,C} = \gamma_{e,C}B$  denote the Larmor frequencies of the spins specified by the indices for the electron and  $^{13}\text{C}$  nuclear spins, with  $\gamma$  being the gyromagnetic ratio.  $A_N = -2.16$  MHz is the secular part of the hyperfine coupling with the  $^{14}\text{N}$  nuclear spin, while  $A_{zz}$  and  $A_{zx}$  are the relevant components of the  $^{13}\text{C}$  hyperfine tensor. We measured  $v_C = 158$ ,  $A_{zz} = -152$ , and  $A_{zx} = 110$  kHz [21]. We chose a weak magnetic field  $B \approx 148$  G, i.e., the electron Larmor frequency  $v_e$  was small compared to the zero-field splitting  $D$ , and far away from the

level anticrossings [45], to simplify the theory model for  $T_1^e$  [46]. Moreover, at this field strength, we have  $|v_C| \approx |A_{zz}|$ , making the system highly suitable for indirect control of the nuclear spin [21,22,47–49].

In the experiment, we used a diamond sample enriched in  $^{12}\text{C}$  and a concentration of substitutional nitrogen less than 10 ppb to achieve long coherence times of the electron spin, which was measured as  $T_2^{*e} = 40$   $\mu\text{s}$  and  $T_2^e = 700$   $\mu\text{s}$  by FID and XY2 dynamical decoupling measurements, respectively [41,42,49–52]. The center is located about 10  $\mu\text{m}$  below the surface of the sample. The experimental setup is presented in the SM (Sec. IV B) [30]. In the following, we write  $|m_S\rangle$  for the eigenstate of  $S_z$  with eigenvalue  $m_S$ , and  $|\uparrow\rangle$  and  $|\downarrow\rangle$  for the eigenstates of  $I_z$  with eigenvalues  $\pm 1/2$ .

To prepare the system for the measurement, we use a combination of laser and MW pulses [21,22] to initialize the system into the state

$$\rho_{\text{ini}}^{e,C} = s_1 |0 \uparrow\rangle \langle 0 \uparrow| + (1 - s_1) |1 \downarrow\rangle \langle 1 \downarrow| \quad (2)$$

with  $s_1 = 0.80$ . The first term is used to generate the nuclear spin coherence. The second term results from the imperfection in polarizing the  $^{13}\text{C}$  spin and contributes observable effects in the detected signals. Details of the pulse sequence for preparing  $\rho_{\text{ini}}^{e,C}$  are given in the SM (Sec. IIB) [30].

### III. EXPERIMENTAL PROTOCOL AND RESULTS

Figure 1 shows the protocol for the relaxation measurements of the  $^{13}\text{C}$  spin as quantum circuits for the free induction decay (FID) (b) and the Hahn echo (c). Here, we only consider the four-level subsystem with the initial state corresponding to the first term in Eq. (2). In this subsystem, we use unitary operations that correspond to a Hadamard gate  $H$  and refocusing operation  $180_x$  for the  $^{13}\text{C}$  spin and the two-qubit SWAP gate. All these operations were implemented without rf pulses, using indirect control for the  $^{13}\text{C}$  spin [21,22]. The final  $90_\phi$  readout pulse converts a component of the electron spin coherence into observable population of the electron spin. Its phase  $\phi$  is incremented in proportion to the free precession time  $\tau$  as

$$\phi = 2\pi \nu_d \tau + \phi_0 \quad (3)$$

to generate an effective offset  $\nu_d$  in the spectrum, such that all resonance lines appear at positive frequencies [42,53,54]. Here  $\nu_d$  and  $\phi_0$  denote the detuning frequency and an initial phase, respectively. In FID and Hahn echo experiments, we set  $\nu_d$  to different values, so that the measured signals have the same frequency, to allow easier comparison, with details as shown in the following. The state of the electron qubit can be detected by laser pulses, as shown in Fig. 1 [4].

Applying the Hadamard gate  $H$  to the  $^{13}\text{C}$  spin generates the superposition state  $|+\rangle = (|\uparrow\rangle + |\downarrow\rangle)/\sqrt{2}$  from  $|\uparrow\rangle$ . Therefore,  $\rho_{\text{ini}}^{e,C}$  shown in Eq. (2) is transformed to

$$\rho_{\text{sup}}^{e,C} = s_1 |0\rangle \langle 0| \otimes |+\rangle \langle +| + (1 - s_1) |1 \downarrow\rangle \langle 1 \downarrow|, \quad (4)$$

where the state of the  $^{13}\text{C}$  spin in the subspace of electron state  $|1\rangle$  remains unchanged by the Hadamard gate  $H$ . We discuss the cases of FID and Hahn echo in the following.

In the FID experiment shown in Fig. 1(b), the nuclear spin coherence is allowed to undergo free evolution

for a time  $\tau$ . The coherence evolves continuously and dephases under the effect of environmental perturbations. These perturbations include the longitudinal relaxation of the electron spin ( $T_1^e$ ): if it flips between states  $|0\rangle$  and  $|-1\rangle$  ( $|1\rangle$ ), the nuclear spin coherence prepared between  $|0, \uparrow\rangle \leftrightarrow |0, \downarrow\rangle$ , with precession frequency  $\nu_0 = \nu_C = 158$  kHz, is transferred to the  $|-1, \uparrow\rangle \leftrightarrow |-1, \downarrow\rangle$  ( $|1, \uparrow\rangle \leftrightarrow |1, \downarrow\rangle$ ) transition, where it precesses with a different frequency  $\nu_{-1} = \sqrt{A_{zx}^2 + (\nu_C + A_{zz})^2} = 110$  kHz [ $\nu_1 = \sqrt{A_{zx}^2 + (\nu_C - A_{zz})^2} = 329$  kHz]. Since this transfer is an incoherent process, the coherence of the nuclear spins coupled to the  $m_S = \pm 1$  electron spin subsystems vanishes.

We denote the populations of the different electron spin levels as  $P_{m_S}(\tau)$  and write the state at the end of the evolution period as

$$\rho_{\text{FID}}^{e,C}(\tau) = \sum_{m_S} P_{m_S}(\tau) (|m_S\rangle\langle m_S|) \otimes \varrho_{\text{FID}}^{C_{m_S}}(\tau), \quad (5)$$

where  $\varrho_{\text{FID}}^{C_{m_S}}(\tau)$  denotes the nuclear spin state in the subspace  $|m_S\rangle$  of the electron spin, and

$$P_0(\tau) = \frac{1}{3} - 2\left(\frac{1}{6} - \frac{s_1}{2}\right)e^{-3\kappa\tau}, \quad (6)$$

$$P_{-1}(\tau) = \frac{1}{3} + \left(\frac{1}{6} - \frac{s_1}{2}\right)e^{-3\kappa\tau} - \frac{1}{2}(1 - s_1)e^{-(\kappa+2\tilde{\kappa})\tau}, \quad (7)$$

$$P_1(\tau) = \frac{1}{3} + \left(\frac{1}{6} - \frac{s_1}{2}\right)e^{-3\kappa\tau} + \frac{1}{2}(1 - s_1)e^{-(\kappa+2\tilde{\kappa})\tau}. \quad (8)$$

Here  $\kappa$  and  $\tilde{\kappa}$  denote the single and double quantum transition rates, respectively, with  $\tilde{\kappa} \approx 2\kappa$  at room temperature [26,55], and  $T_1^e = 1/(3\kappa)$ , where its experimental value is 5.5 ms. Details are in the SM (Sec. I) [30]. The state of the nuclear spin in the electron spin subspace  $|1\rangle$  does not contribute to the observed signals, since the readout is selective for the subspace  $|0\rangle, |-1\rangle$ . In the subspace  $m_S = 0$  the nuclear spin state is

$$\varrho_{\text{FID}}^{C_0}(\tau) = \frac{1}{2} \begin{pmatrix} 1 & c_{\text{FID}}(\tau)e^{-i2\pi\nu_C\tau} \\ c_{\text{FID}}^*(\tau)e^{i2\pi\nu_C\tau} & 1 \end{pmatrix}. \quad (9)$$

In the following, we assume an exponential decay of the coherence  $c_{\text{FID}}(\tau) = e^{-\tau/T_2^C}$ . More details on Eq. (9) are presented in the SM (Sec. III A) [30]. The populations  $P_{\pm 1}(\tau)$  of the electron spin state  $|\pm 1\rangle$  are the result of incoherent ( $T_1^e$ ) process and therefore no nuclear spin coherence builds up in these subsystems.

To generate an observable signal from the nuclear spin coherence, we use a three-step process: we first apply a SWAP operation to the two-qubit system to convert the nuclear spin coherence into electron spin coherence; then we apply a  $90^\circ$  pulse to the electron and count the fluorescence photons during the readout-laser pulse [4].

The SWAP operation exchanges the states of the  $^{13}\text{C}$  nuclear spin qubit with that of the electron spin qubit. It transforms the state  $\rho_{\text{FID}}^{e,C}(\tau)$  in Eq. (5) to

$$\begin{aligned} \rho_{\text{FID},S}^{e,C}(\tau) &= P_0(\tau)\varrho_{\text{FID}}^{C_0}(\tau) \otimes (|\uparrow\rangle\langle\uparrow|) \\ &+ P_{-1}(\tau)E/2 \otimes (|\downarrow\rangle\langle\downarrow|) \\ &+ P_1(\tau)|1\rangle\langle 1| \otimes \varrho_{\text{FID}}^{C_1}(\tau). \end{aligned} \quad (10)$$

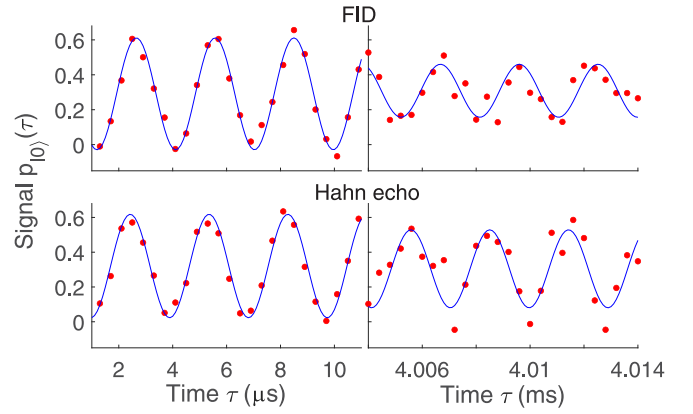


FIG. 2. Measured signals with the fitted curves for the FID and Hahn echo.

Here,  $\varrho_{\text{FID}}^{C_0}(\tau)$  is defined in Eq. (9) and  $E$  denotes the  $2 \times 2$  unit operator. The SWAP operation has therefore transferred the coherence  $c_{\text{FID}}(\tau)$  to the electron spin states. From there, it is converted into a population difference by a  $90^\circ$  MW pulse with the phase  $\phi$  shown in Eq. (3). Finally, a readout laser pulse measures the population of the electron spin in the state  $|0\rangle$ ,

$$p_{|0\rangle,\text{FID}}(\tau) = (1/2) \{ [P_0(\tau) + P_{-1}(\tau)] + P_0(\tau)c_{\text{FID}}(\tau)\sin(2\pi\nu_s\tau + \phi_0) \}, \quad (11)$$

where  $\nu_s = -(\nu_C + \nu_d^a) = 342$  kHz, with detuning frequency  $\nu_d^a = -0.5$  MHz. Here and in the following  $p$  refers to the population after the SWAP operation and the  $90^\circ$  pulse and  $P$  to the population before the SWAP. Figure 2 illustrates the measured  $p_{|0\rangle,\text{FID}}(\tau)$  for two time segments. More results are shown in the SM (Sec. III C) [30].

The signal  $p_{|0\rangle,\text{FID}}(\tau)$  in Eq. (11) consists of an oscillating part that reflects the coherence in the system, with amplitude  $P_0(\tau)c_{\text{FID}}(\tau)/2$  and a background  $[P_0(\tau) + P_{-1}(\tau)]/2$ , which reflects the mean of the populations. Figure 3 shows the experimental results, where we fit the measured background in

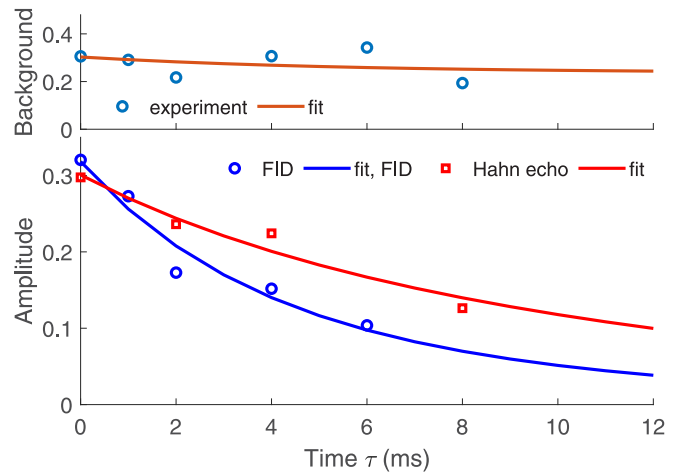


FIG. 3. Experimental results for the decay of the nuclear spin coherence. The upper part shows the background and the lower part the coherence amplitudes for the FID and Hahn echo experiments.

TABLE I. Parameters obtained by fitting the experimental data in Fig. 3 to the function (13) or (17), with standard errors. The details on the normalization of the experimental data are presented in the SM (Sec. II) [30].

|                             |                                     |                             |
|-----------------------------|-------------------------------------|-----------------------------|
| $^{13}\text{C}$ , FID       | $c_{0,\text{FID}} = 0.80 \pm 0.05$  | $T_2^{*C} = 8.7 \pm 2.4$ ms |
| $^{13}\text{C}$ , Hahn echo | $c_{0,\text{Hahn}} = 0.75 \pm 0.04$ | $T_2^C = 12.7 \pm 2.9$ ms   |

$p_{|0\rangle,\text{FID}}(\tau)$  to the function

$$[P_0(\tau) + P_{-1}(\tau)]/2 - d_0 \quad (12)$$

with the offset  $d_0 = 0.086$ . More details are presented in the SM (Sec. III D) [30]. By fitting the measured amplitude of the coherence  $p_{|0\rangle,\text{FID}}(\tau)$  to the function  $P_0(\tau)c_{\text{FID}}(\tau)/2$ , where

$$c_{\text{FID}}(\tau) = c_{0,\text{FID}}e^{-\tau/T_2^{*C}}, \quad (13)$$

we obtain the dephasing time  $T_2^{*C}$ . The experimental results and the corresponding fits are shown in Fig. 3 (bottom), where the amplitude denotes  $P_0(\tau)c_{\text{FID}}(\tau)/2$  or  $[P_0(\tau) + P_{-1}(\tau)]c_{\text{Hahn}}(\tau)/2$  for the FID or Hahn echo experiment (see the following), respectively. In Table I, we list the obtained parameters.

The Hahn echo experiment starts with the same initialization as the FID experiment but uses an inversion operation in the middle of the evolution period to partly refocus the evolution, as shown in Fig. 1(c). The state at the qubit system at the end of the full evolution ( $\frac{\tau}{2} - 180_x - \frac{\tau}{2}$ ) and the SWAP operation is

$$\begin{aligned} \rho_{\text{Hahn},S}^{e,C}(\tau) &= \mathcal{Q}_{\text{Hahn}}(\tau) \otimes \begin{pmatrix} P_0(\tau) & 0 \\ 0 & P_{-1}(\tau) \end{pmatrix} \\ &+ P_1(\tau)|1\rangle\langle 1| \otimes \mathcal{Q}_{\text{Hahn}}^C(\tau), \end{aligned} \quad (14)$$

where the operator

$$\mathcal{Q}_{\text{Hahn}}(\tau) = \frac{1}{2} \begin{pmatrix} 1 & c_{\text{Hahn}}(\tau) \\ c_{\text{Hahn}}(\tau) & 1 \end{pmatrix}, \quad (15)$$

with  $c_{\text{Hahn}}(\tau) = e^{-\tau/T_2^C}$ , is now an electron spin operator in the ( $m_S = 0, m_S = -1$ ) subspace. By comparing Eqs. (15) and (9), we can see that the inversion operation in the Hahn echo experiment has refocused the nuclear precession with frequency  $\nu_C$ . Additional details are given in the SM (Sec. IIIA) [30]. Similar to the detection in the FID experiment, we apply a  $90^\circ$  MW pulse with the phase  $\phi$  to obtain the population

$$\begin{aligned} p_{|0\rangle,\text{Hahn}}(\tau) &= [P_0(\tau) + P_{-1}(\tau)]/2 \\ &\times [1 + c_{\text{Hahn}}(\tau) \sin(2\pi\nu_d^b\tau + \phi_0)]. \end{aligned} \quad (16)$$

Here we choose the detuning frequency as  $\nu_d^b = -342$  kHz, different from the case of the FID, so that the resulting oscillation frequencies  $\nu_s$  are the same. The experimental data can then be fitted to  $[P_0(\tau) + P_{-1}(\tau)]c_{\text{Hahn}}(\tau)/2$ , where

$$c_{\text{Hahn}}(\tau) = c_{0,\text{Hahn}}e^{-\tau/T_2^C}. \quad (17)$$

This allows us to extract  $T_2^C$ . Unlike the case of the FID experiment, to extract  $T_2^C$  from the Hahn echo experiment we need the value for  $\tilde{\kappa}$ , as evidenced by comparing Eqs. (11) and (16). Here we set  $\tilde{\kappa} = 2\kappa$  [26,55] and discuss the related uncertainty in the SM (Sec. III E) [30]. Table I lists the resulting

parameters. They show that the refocusing operation in the Hahn echo extends the dephasing by 50% compared to the FID.

#### IV. DISCUSSION

Here, we only used a single echo of the nuclear spin in a subspace of two states  $|0\rangle$  and  $|-1\rangle$  of the electron spin. The effect of electron spin flips between states  $|0\rangle$  and  $|-1\rangle$  due to  $T_1^e$  relaxation can be suppressed in the following way. In the subspace  $|0\rangle$ , the echo refocuses the evolution of the nuclear spin, while in subspace  $|-1\rangle$ , the nuclear spin can be approximated by an eigenstate of the effective Hamiltonian ( $\sim I_x$ ) and therefore does not evolve. We present more details in the SM (Sec. III) [30]. Since we do not generate an echo to the nuclear spin for the electron state  $m_S = 1$ , flips between  $m_S = 0$  and  $m_S = 1$  still cause nuclear dephasing. Therefore, from Eq. (16), one finds that  $T_1^e$  still limits the amplitude of the detected signals in measuring the coherence of the nuclear spin, although the effect is smaller than in the case of the FID experiment; see Eq. (11). We believe that multiple echos in the whole electron space should extend the coherence time further [56,57].

#### V. CONCLUSION AND OUTLOOK

We have measured the dephasing times of the  $^{13}\text{C}$  nuclear spin in a single NV center during free precession and with partial refocusing in a Hahn echo experiment. We specifically worked with a system with strong hyperfine couplings between the electron and nuclear spins. Compared with previous works, e.g., Refs. [12,13,15,58], we introduced an improved detection scheme for the nuclear spin coherence, which relies on a SWAP gate and we did not use additional laser pulses that might depolarize and dephase the nuclear spins. Our results demonstrate that the nuclear spin coherence time can exceed the longitudinal relaxation time of the electron spin in a hybrid spin system. In the case of the FID, the nuclear spin dephasing time  $T_2^{*C}$  exceeds  $T_1^e$  by a factor of 1.6. Using one refocusing operation, we further extended the coherence time to 2.3 times  $T_1^e$ .

In quantum information processing, the nuclear spins can act as memory qubits. Therefore, our results are useful for extending the quantum memory times of quantum computers. Increasing the polarization of  $^{14}\text{N}$  could further improve the experiment, since more population of the chosen subsystem yields a stronger signal. Our results might provide a useful method to investigate the physical mechanism for electron spin relaxation ( $T_1^e$ ) [59], using the nuclear spin as an ancillary quantum memory or a probe qubit [17,60,61] to measure the frequency distribution of the noise from  $T_1^e$  by applying dynamical decoupling pulses [62–64]. This should be applicable in systems with similar mechanisms of nuclear dephasing, such as the stochastic process from the laser reinitialization on the nuclear qubits in quantum networks [14,16]. Our results are obtained in a system with stronger couplings than previous works, which might be useful for improving the communication efficiency in quantum networks. In general, stronger couplings lead to shorter gate operations for nuclear spin qubits [23]. Our experiment was performed in NV centers. The method should also work in other systems, such as malonic acid radical [48,65] and other defects in solids [19,66].

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## DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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