Experimental analysis of noise strength and environmental correlation time for ensembles of nitrogen-vacancy centers in diamond

K. Hayashi¹, Y. Matsuzaki², T. Ashida¹, S. Onoda³, H. Abe³, T. Ohshima³, M. Hatano⁴, T. Taniguchi⁵, H. Morishita¹, M. Fujiwara¹, N. Mizuochi¹

¹Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan
²Nanoelectronics Research Institute, National Institute of Advanced Industrial Science and Technology (AIST), 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568, Japan.
³National Institutes for Quantum and Radiological Science and Technology (QST), Takasaki, Gunma 370-1292, Japan
⁴Department of Physical Electronics, Tokyo Institute of Technology, 2-12-1 Ookayama, Meguro-ku, Tokyo 152-8552, Japan
⁵National Institute for Materials Science (NIMS), Tsukuba, Ibaraki 305-0044, Japan

Abstract

We experimentally and theoretically investigate the Hahn echo decay curve for nitrogen vacancy centers in diamond with different spin concentrations. The Hahn echo results show a non-exponential decay for low spin concentrations, while an exponential decay is dominant for high spin concentrations. By fitting the decay curve with a theoretical model, we show that both the amplitude and correlation time of the environmental noise have a clear dependence on the spin concentration. These results are essential for optimizing the NV center concentration in high-performance quantum devices, particularly quantum sensors.
Nitrogen vacancy centers (NV centers) in diamond are a promising system for realizing quantum information processing [1-9,16–35]. An NV center is a spin 1 system. Magnetic fields parallel to the NV center axis lift the degeneracy between $m_s = +1$ and $m_s = -1$ by a Zeeman shift $\Delta E = \gamma B$, where $\gamma$ is the gyromagnetic ratio for the electron spin (28 MHz/mT) and $B$ is the magnetic field parallel to the NV center axis. Microwaves with a frequency of around 2.87 GHz can induce a transition between $m_s = 0$ and $m_s = \pm 1$ for the electronic ground state. By using frequency selectivity, we can use this system as an effective two-level system. The electronic ground state for the NV center spin ($S = 1$) can be initialized and read by optical pumping, allowing us to perform optically detected magnetic resonance (ODMR) with NV centers [1]. Moreover, NV centers have a long coherence time and stability even at room temperature [2]. These properties indicate NV centers have the potential to realize quantum information processing involving quantum memory [3–6], quantum simulation [7–9], and distributed quantum computation [10–15]. One of the most attractive applications of NV centers is for high sensitivity magnetometers [16–28]. A single NV center allows nanoscale magnetometry with a reasonably high sensitivity ($\sim$ nT/Hz$^{1/2}$) [2,16,17], making it possible to perform nano-NMR [18]. For a sensor with a size of about a micron, where magnetoencephalography and magnetocardiogram applications are possible, an ensemble of NV centers has a high sensitivity proportional to the square root of the number of NV centers [19–23]. A sensitivity of 0.9 pT/Hz$^{1/2}$ has been demonstrated with $1.4 \times 10^{11}$ NV centers using spin echo based magnetometry [19]. Also, several techniques such as the use of a sophisticated pulse sequence [25–28] or hybridization with other systems [3–6] have expanded the potential of NV based magnetometers. Furthermore, theoretical calculations have shown that the optimal sensitivity is around 250 aT/$\sqrt{\text{Hz}}$/cm$^{2/3}$, which is comparable to that of SQUIDs [16], though the sensitivity of NV magnetometers is currently lower than the optimal value and still needs to be improved.

Importantly, NV centers suffer from decoherence, which is an obstacle for more practical applications. By applying a $\pi/2$-pulse, we can create a superposition between $m_s = 0$ and $m_s = \pm 1$. However, the coherent superposition of two states accumulates a relative phase of the target magnetic field, and low frequency fluctuations of the magnetic field induce dephasing of the NV centers. Importantly, a spin echo pulse sequence eliminates such environmental noise and thus improves the coherence time [2,16,17]. This technique is widely used in quantum information processing with NV centers [2,16,17]. With a high-density ensemble of NV centers, dipole interactions from nitrogen paramagnetic impurities (P1 centers) are a dominant noise source for spin dephasing [16,17,29–33]. Interactions between the NV centers are another strong noise source [16,33]. To realize a practical sensor, it is essential to understand how the nitrogen concentration affects decoherence, which will let us determine a strategy to improve the coherence time. There are many ways to suppress decoherence, such as the quantum Zeno effect.
dynamical decoupling [29, 44, 45], qubit motion [46, 47], and quantum error correction [39–42]. However, the efficiency of these protocols strongly depends on the noise properties. For example, the quantum Zeno effect and qubit motion can be used when quadratic decay is experimentally observed, while these schemes do not improve the coherence time for exponentially decaying systems. Dynamical decoupling can be used to suppress decoherence only when the environment is non-Markovian. Quantum error correction can detect noise and recover quantum states for any type of local decoherence [47, 48]. However, when using NV centers for quantum sensors, quantum error correction could suppress not only decoherence but also signal accumulation from the target fields, and so a careful assessment of the environment and choice of error correction code is necessary in order to improve the sensitivity when using quantum error correction [39–42].

Also, when using NV centers for entanglement enhanced magnetic field sensors, it is crucial to understand the origin of decoherence [49, 50]. Recently, a systematic way to create a few NV centers within a distance of tens of nanometers has been developed [51], which paves the way to realizing a sensitive quantum sensor with entangled NV centers. However, from a theoretical point of view, it is known that an entanglement sensor is more effective than a classical sensor (that uses separable states) only when the decoherence behavior shows non-exponential decay [49, 50]. This means that for practically useful entanglement sensors, understanding and controlling the environment is necessary.

In this article, we report a systematic study of Hahn echo decay curves for NV centers in several diamond samples with different NV center concentrations. Our experimental results show that an exponential decay curve is observed for a high nitrogen concentration and a non-exponential decay curve is observed for a low nitrogen concentration. Using our model, we can fit the decay curve for both low and high NV center concentrations. Our analysis also shows that the correlation time and amplitude of the dipole interaction from the environmental spin bath have a linear dependence on the spin concentration.

Nine sample diamond crystals with substitutional nitrogen (P1) centers were used in our study. The diamonds were synthesized by high-pressure high-temperature (HPHT) processes [52]. Among them, samples No. 5–9 were commercially obtained. The NV centers were created through 2-MeV electron irradiation at various doses (the irradiation doses are shown in Table 1) at 745±10 °C and subsequent annealing at 1000 °C for 1 hour in Ar gas. After annealing, the diamonds were cleaned in a mixture of boiling acids (1:1 sulfuric, nitric) to remove graphitic carbon and oxygen-termination at the surface. The P1 center concentrations were evaluated by electron paramagnetic resonance (EPR) spectroscopy at room temperature after electron irradiation and annealing. The NV center concentrations were investigated by comparing the fluorescent intensity between a single NV center and our samples. The fluorescence intensity was
measured by applying a 532-nm laser with a lab-built confocal microscope (detection volume: 0.04 \, \mu \text{m}^3). Table 1 shows both the P1 and NV concentrations for the nine diamond samples, estimated from the EPR and fluorescence measurements. The Hahn echo sequence (Fig. 1(a)) for the diamonds was performed at room temperature. Green laser pulses at 50 \, \mu\text{W} illuminated the diamond samples. Microwave pulses were applied by a copper wire above the diamond surface. A magnetic field (~30 mT) was applied along the <111> direction using a permanent magnet to separate the revival peaks from the Hahn echo decay.

Next, we fit our experimental results using an analytical expression for the decay curves. First, we fit the results with a simple stretched exponential function \( \exp(-\frac{t}{\tau_2})^n \) and estimated the coherence time \( T_2 \). Fig. 1(b) shows an almost linear dependence on the spin concentration (the sum of the NV center concentration and P1 center concentration). Although the stretched exponential function is useful for estimating \( T_2 \) of the NV center, this fit does not capture some important properties of the environment. So, we adopted a random classical Gaussian noise model that contains the noise amplitude and correlation time of the environment, as we will describe later. This model is used to study the noise properties of the spin bath of a single NV center [29]. We introduce the decoherence of a single qubit when we perform a spin echo [16]. The interaction Hamiltonian between the qubit and the environment is considered to be

\[
H(t) = \lambda f(t) \hat{\sigma}_z.
\]

We consider non-biased noise such that \( \overline{f(t)f(0)} = 0 \). From this model, we can derive the Hahn echo decay curve as (see the appendix for details):

\[
\langle 1 | \rho_t(\tau) | 0 \rangle = \frac{1}{2} e^{-\lambda t^2 \tau_c} \left( t_c + \frac{\tau}{4} e^{-\frac{\tau}{\tau_c}} \right) \left( 3 e^{-\frac{\tau}{\tau_c}} + 4 e^{-\frac{2\tau}{\tau_c}} \right)
\]

For \( \tau \ll \tau_c \), we have \( \langle 1 | \rho_t(\tau) | 0 \rangle \approx \frac{1}{2} e^{-\frac{\lambda \tau^2}{\tau_c^2}} \), while we have \( \langle 1 | \rho_t(\tau) | 0 \rangle \approx \frac{1}{2} e^{-\lambda \tau \tau_c} \) for \( \tau \gg \tau_c \). These results are consistent with previous work [10]. In Fig. 3, we show the fitting results for
the Hahn echo decay curves. We observed periodic revival peaks due to rephasing of the $^{13}$C spin bath [53,54]. Since we are interested in the decoherence effect from an electron spin bath that induces a monotonic decay, we performed a fit of the envelope of the data. The theoretical calculations show that we can observe a non-exponential decay for a long correlation time while an exponential decay should be dominant for a short correlation time. Actually, our experiments show a non-exponential decay with lower density samples while the decay becomes exponential for higher density samples. Intuitively, this shows that if the density of the electron spin is increased, the correlation time should be shorter. For a more quantitative analysis, we plot the values of $\tau_c$ and $\lambda$, obtained from fitting our experimental data using Eq. (10), with respect to the spin concentration (the sum of the NV center concentration and P1 center concentration) in Fig. 3. These parameters show an almost linear dependence on the spin concentration. Note that this is the first systematic and quantitative investigation of how the Hahn echo decay curve depends on the spin concentration.

Finally, we discuss possible future work. Regarding fabrication, there is room to improve the coherence time for the ensemble of NV centers. Especially, our HPHT samples contain some residual metal impurities that decrease the coherence time for the NV centers. Moreover, by decreasing the $^{13}$C nuclear spins, we could eliminate unwanted oscillations in the spin echo signal. For our theoretical model, where we have adopted a semi-classical noise model, we could instead use a microscopic model that can partially include many-body quantum effects [53]. While our semi-classical model can successfully reveal a clear relation between the noise parameters and spin concentration, a fully quantized model could give a deeper understanding of the decoherence mechanism for the NV centers.

We have investigated the Hahn echo decay curve for nine samples of ensembles of NV centers with different spin concentrations. Our results show a crossover from an exponential decay for high density samples to a non-exponential decay for low density samples. Our theoretical analysis of these experimental data reveals that the crossover between the forms of the decay is due to a change in the noise amplitude and environmental correlation time. By fitting the data with a theoretical decay curve, we show that the noise amplitude and the environmental correlation time have a linear dependence on the spin concentration of the diamond sample. Our results are crucial for understanding the decoherence dynamics of NV centers in diamond with various spin densities and will be useful for developing applications of NV centers for quantum information processing.

ACKNOWLEDGMENTS

This work was supported by the Leading Initiative for Excellent Young Researchers MEXT
Japan, and in part supported by MEXT KAKENHI (Grant Nos. 15H05868, 15H05870, 16H06326), CREST (JPMJCR1333) and MEXT Q-LEAP.

APPENDIX

We solve the Schrodinger equation in an interaction picture to obtain

$$\rho_t(\tau) = \rho(0) - i \int_0^\tau dt [H_t(t_1), \rho(t_1)]$$

$$= \rho(0) + \sum_{n=1}^\infty (-i)^n \int_0^\tau \cdots \int_0^\tau dt_1 dt_2 \cdots dt_n [H_t(t_1), \cdots, [H_t(t_n), \rho(t_0)]]$$

By taking the average, we obtain

$$\overline{\rho_t(\tau)} = \rho(0) + \sum_{n=1}^\infty (-i\lambda)^n \frac{1}{n!} \int_0^\tau \cdots \int_0^\tau dt_1 dt_2 \cdots dt_n \left[ \begin{array}{c} f'(t_1) \cdots f'(t_{2m}) \end{array} \right]$$

For a random Gaussian variable, all the cumulants except the second-order terms tend to be zero.

Thus, we can ignore all moments of odd order, giving

$$f'(t_1) \cdots f'(t_{2m}) = (2m-1)(2m-3) \cdots 1 \left( f'(t_1) f'(t_2) \right)^m$$

So, we obtain

$$\overline{\rho_t(\tau)} = \rho(0) + \sum_{m=1}^\infty (-i\lambda)^{2m} \frac{1}{m!} \left( \frac{1}{2m} \int_0^\tau dt_1 dt_2 \left( \overline{f(t_1) f(t_2)} g(t_1) g(t_2) \right)^m \left[ \begin{array}{c} \hat{\sigma}_z, \hat{\sigma}_z, \cdots, [\hat{\sigma}_z, \rho(0)] \end{array} \right] \right)$$

Here, we assume $\overline{f(t_1) f(t_2)} = e^{\frac{t_1 - t_2}{\tau_c}}$, and we obtain

$$\int_0^\tau dt_1 dt_2 \left( e^{\frac{t_1 - t_2}{\tau_c}} g(t_1) g(t_2) \right) = \int_0^\tau dt_1 dt_2 \left( e^{\frac{t_1 - t_2}{\tau_c}} g(t_1) g(t_2) \right)$$
\begin{equation}
\tau_c \chi + \left( -3 - e^{-\frac{\chi}{\tau_c}} + 4e^{-\frac{2\chi}{2\tau_c}} \right)(\tau_c)^2
\end{equation}

Finally, we obtain
\begin{equation}
\rho_i(\tau) = \rho(0) + \sum_{m=1}^{\infty} \frac{(-i\lambda)^{2m}}{m!} \left( \tau_c \chi + \left( -3 - e^{-\frac{\chi}{\tau_c}} + 4e^{-\frac{2\chi}{2\tau_c}} \right)(\tau_c)^2 \right)^m \left[ \hat{\sigma}_z, [\hat{\sigma}_z, \ldots, [\hat{\sigma}_z, \rho(0)] \right]_{2m}
\end{equation}

For an initial state $|+\rangle = \frac{1}{\sqrt{2}} (|0\rangle + |1\rangle)$, the non-diagonal terms of the density matrix show the decay curve, and we describe the Hahn echo decay curve as

\begin{equation}
\langle 1 | \rho_i(\tau) | 0 \rangle = \frac{1}{2} e^{-4\lambda^2 \left( \tau_c \chi + \left( -3 - e^{-\frac{\chi}{\tau_c}} + 4e^{-\frac{2\chi}{2\tau_c}} \right)(\tau_c)^2 \right)}
\end{equation}

REFERENCE

[1] A. Gruber, A. Dräbenstedt, C. Tietz, L. Fleury, J. Wrachtrup, and C. Von Borczyskowski, Science 276, 2012 (1997).
[2] G. Balasubramanian, P. Neumann, D. Twitchen, M. Markham, R. Kolesov, N. Mizuochi, J. Isoya, J. Achard, J. Beck, J. Tissler, V. Jacques, P. R. Hemmer, F. Jelezko, and J. Wrachtrup, Nat. Mater. 8, 383 (2009).
[3] X. Zhu, S. Saito, A. Kemp, K. Kakuyanagi, S. Karimoto, H. Nakano, W. J. Munro, Y. Tokura, M. S. Everitt, K. Nemoto, M. Kasu, N. Mizuochi and K. Semba, Nature 478, 221 (2011).
[4] S. Saito, X. Zhu, R. Amsüss, Y. Matsuzaki, K. Kakuyanagi, T. Shimo-Oka, N. Mizuochi, K. Nemoto, W. J. Munro, and K. Semba, Phys. Rev. Lett. 111, 107008 (2013).
[5] M. Yuichiro, X. Zhu, K. Kakuyanagi, H. Toida, T. Shimo-Oka, N. Mizuochi, K. Nemoto, K. Semba, W. J. Munro, H. Yamaguchi, and S. Saito, Phys. Rev. Lett. 114, 120501 (2015).
[6] Y. Kubo, C. Grezes, A. Dewes, T. Umeda, J. Isoya, H. Sumiya, N. Morishita, H. Abe, S. Onoda, T. Oshshima, V. Jacques, A. Dréau, J.-F. Roch, I. Diniz, A. Auffeves, D. Vion, D. Esteve, and P. Bertet, Phys. Rev. Lett. 107, 220501 (2011).
[7] J. Choi, S. Choi, G. Kucsko, P. C. Maurer, B. J. Shields, H. Sumiya, S. Onoda, J. Isoya, E. Demler, F. Jelezko, N. Y. Yao, and M. D. Lukin, Phys. Rev. Lett. 118, 093601 (2017).
[8] S. Choi, J. Choi, R. Kandig, G. Kucsko, H. Zhou, J. Isoya, F. Jelezko, S. Onoda, H. Sumiya, V. Khemani, C. von Keyserlingk, N. Y. Yao, E. Demler and M. D. Lukin, Nature, 543, 221 (2017).
[9] E. Bauch, C. A. Hart, J. M. Schloss, M. J. Turner, J. F. Barry, P. Kehayias, S. Singh, and R. L. Walsworth, Phys. Rev. X 8, 031025 (2018).
[10] C. Cabrillo, J. I. Cirac, P. García-Fernández, and P. Zoller, Phys. Rev. A 59, 1025 (1999).
[11] S. Bose, P. L. Knight, M. B. Plenio, and V. Vedral, Phys. Rev. Lett. 83, 5158 (1999).
[12] C. W. Chou, H. de Riedmatten, D. Felinto, S. V. Polyakov, S. van Enk, and H. J. Kimble, Nature 438, 828 (2005).
[13] D. L. Moehring, P. Maunz, S. Olmschenk, K. C. Younge, D. N. Matsukevich, L.-M. Duan and C. Monroe, Nature (London) 449, 68 (2007).
[14] Y. Lim, A. Beige, and L. Kwok, Phys. Rev. Lett. 95, 030505 (2005).
[15] S. D. Barrett and P. Kok, Phys. Rev. A 71, 060310(R) (2005).
[16] J. M. Taylor, P. Cappellaro, L. Childress, L. Jiang, D. Budker, P. R. Hemmer, A. Yacoby, R. Walsworth, and M. D. Lukin, Nat. Phys. 4, 810 (2008).
[17] J. R. Maze, P. L. Stanwix, J. S. Hodges, S. Hong, J. M. Taylor, P. Cappellaro, L. Jiang, M. V. Gurudev, D. E. Togan, A. S. Zibrov, A. Yacoby, R. L. Walsworth, and M. D. Lukin, Nature 455, 644 (2008).
[18] N. Aslam, M. Pfender, P. Neumann, R. Reuter, A. Zappe, F. Favaro de Oliveira, A. Denisenco, H. Sumiya, S. Onoda, J. Isoya, and J. Wrachtrup, Science 357, 67 (2017).
[19] T. Wolf, P. Neumann, K. Nakamura, H. Sumiya, T. Ohshima, J. Isoya, and J. Wrachtrup, Phys. Rev. X 5, 041001 (2015).
[20] D. Duan, G. X. Du, V. K. Kavatamane, S. Arumugam, Y. -K. Tzeng, H-C Chang, and G. Balasubramanian, Opt. Express 27, 6734 (2019).
[21] V. M. Acosta, E. Bauch, M. P. Ledbetter, C. Santori, K. -M. C. Fu, P. E. Barclay, R. G. Beausoleil, H. Linget, J. F. Roch, F. Treussart, S. Chemerisov, W. Gawlk, and D. Budker, Phys. Rev. B 80, 115202 (2009).
[22] J. F. Barry, M. J. Turner, J. M. Schloss, D. R. Glenn, Y. Song, M. D. Lukin, H. Park, and R. L. Walsworth, Proc. Natl. Acad. Sci. U. S. A. 113, 14133 (2016).
[23] Y. Dumeige, M. Chipaux, V. Jacques, F. Treussart, J.-F. Roch, T. Debuisschert, V. M. Acosta, A. Jarmola, K. Jensen, P. Kehayias, and D. Budker, Phys. Rev. B 87, 155202 (2013).
[24] A. Ajoy, U. Bissbort, M.D. Lukin, R. L. Walsworth, and P. Cappelaro, Phys. Rev. X 5, 011001 (2015).
[25] A. Lazariiev, S. Arroyo-Camejo, G. Rahane, V. K. Kavatamane, and G. Balasubramanian, Sci. Rep. 7, 6586 (2017).
[26] S. Schmitt, T. Gefen, F. M. Stürner, T. Unden, G. Wolff, C. Müller, J. Scheuer, B. Naydenov, M. Markham, S. Pezzagna, J. Meijer, I. Schwarz, M. Plenio, A. Retzker, L. P. McGuinness, and F. Jelezko, Science 356, 832 (2017).
[27] J. M. Boss, K. S. Cujia, J. Zopes, and C. L. Degen, Science 356, 837 (2017).
[28] B. Naydenov, F. Dolde, L. T. Hall, C. Shin, H. Fedder, L. C. L. Hollenberg, F. Jelezko, and J. Wrachtrup, Phys. Rev. B 83, 081201(R) (2011).
[29] G. de Lange, Z. H. Wang, D. Riste, V. V. Dobrovitski, and R. Hanson, Science 330, 60 (2010).
[30] K. Hayashi, Y. Matsuzaki, T. Taniguchi, T. Shimo-Oka, I. Nakamura, S. Onoda, T. Ohshima, H. Morishita, M. Fujiwara, S. Saito, and N. Mizuochi, Phys. Rev. Appl. 10, 034009 (2018).
[31] R. Hanson, V. V. Dobrovitski, A. E. Feiguin, O. Gywat, and D. D. Awschalom, Science 320, 352 (2008).
[32] R. Hanson, F. M. Mendoza, R. J. Epstein, and D. D. Awschalom, Phys. Rev. Lett. 97, 087601 (2006).
[33] G. Kucsko, S. Choi, J. Choi, P. C. Maurer, H. Zhou, R. Landig, H. Sumiya, S. Onoda, J. Isoya, F. Jelezko, E. Demler, N. Y. Yao, and M. D. Lukin, Phys. Rev. Lett. 121, 023601 (2018).
[34] W. M. Itano, D. J. Heinzen, J. J. Bollinger, and D. J. Wineland, Phys. Rev. A 41, 2295 (1990).
[35] J. Wolters, M. Strauß, R. S. Schoenfeld, and O. Benson, Phys. Rev. A 88, 020101(R) (2013).
[36] N. Kalb, J. Cramer, J. Twitchen, M. Markham, R. Hanson, and T. H. Taminiau, Nat. Commun. 7, 13111 (2016).
[37] T. Kondo, Y. Matsuzaki, K. Matsushima, and J. G. Filgueiras, New J. Phys. 18, 013033 (2016).
[38] Y. Matsuzaki, T. Shimo-Oka, H. Tanaka, Y. Tokura, K. Semba, and N. Mizuochi, Phys. Rev. A 94, 052330 (2016).
[39] E. M. Kessler, I. Lovchinsky, A. O. Sushkov, and M. D. Lukin, Phys. Rev. Lett. 112, 150802 (2014).
[40] G. Arrad, Y. Vinkler, D. Aharonov, and A. Retzker, Phys. Rev. Lett. 112, 150801 (2014).
[41] W. Dür, M. Skotiniotis, F. Fröwis, and B. Kraus, Phys. Rev. Lett. 112, 080801 (2014).
[42] D. A. Herrera-Martí, T. Gefen, D. Aharonov, N. Katz, and A. Retzker, Phys. Rev. Lett. 115, 200501 (2015).
[43] Y. Matsuzaki, and S. Benjamin, Phys. Rev. A 95, 032303 (2017).
[44] N. Bar-Gill, L. M. Pham, A. Jarmola, D. Budker, and R. L. Walsworth, Nat. commun. 4, 1743 (2013).
[45] B. Naydenov, F. Dolde, L. T. Hall, C. Shim, H. Fedder, L. C. L. Hollenberg, F. Jelezko, and J. Wrachtrup, Phys. Rev. B 83, 081201 (2011).
[46] Y. Matsuzaki, S. Benjamin, S. Nakayama, S. Saito, and W. J. Munro, Phys. Rev. lett. 120, 140501 (2018).
[47] D. V. Averin, K. Xu, Y. P. Zhong, C. Song, H. H. Wang, and S. Han, Phys. Rev. Lett. 116, 010501 (2016).
[48] R. Laflamme, C. Miquel, J. P. Paz, and W. H. Zurek, Phys. Rev. Lett. 77, 198 (1996).
[49] Y. Matsuzaki, S. C. Benjamin, and J. Fitzsimons, Phys. Rev. A 84, 012103 (2011).
[50] A. W. Chin, S. F. Huelga, and M. B. Plenio, Phys. Rev. lett. 109, 233601 (2012).
[51] M. Haruyama, S. Onoda, T. Higuchi, W. Kada, A. Chiba, Y. Hirano, T. Teraji, R. Igarashi, S. Kawai, H. Kawarada, Y. Ishii, R. Fukuda, T. Tanii, Y. Isoya, T. Ohshima, and O. Hanaizumi, Nat. Common. 10, 2664 (2019).
[52] H. Kanda, Braz. J. of Phys. 30, 482 (2000).
[53] L. Childress, M. V. Gurudev Dutt, J. M. Taylor, A. S. Zibrov, F. Jelezko, J. Wrachtrup, P. R. Hemmer, and M. D. Lukin, Science 314, 281 (2006).
[54] P. Huang, X. Kong, N. Zhao, F. Shi, P. Wang, X. Rong, R. Liu, and J. Du, Nat. Common. 2, 570 (2011).
Fig. 1. (a) Typical Hahn echo sequence. (b) $T_2$ estimated by fitting experimental results with stretched exponential decay curve.
Fig. 2. Fitting results for Hahn echo decay curve for all diamond samples. The black and red lines are the experimental and numerical results, respectively. Eq. (10) was used for the fitting. Suitable magnetic fields were applied in the diamonds $<111>$ direction.

Fig. 3. Plots of (a) $\lambda$ and (b) $1/t_c$ with respect to spin concentration. The parameters were estimated by
fitting the experimental results using Eq. (10).

Table 1

|   | P1 concentration ($\times 10^{17} / \text{cm}^3$) | NV concentration ($\times 10^{17} / \text{cm}^3$) | Electron irradiation dose ($\times 10^{16} \text{e/cm}^2$) |
|---|---------------------------------|---------------------------------|---------------------------------|
| No. 1 | 33.6 | 18.4 | 100 |
| No. 2 | 11.6 | 11.2 | 50 |
| No. 3 | 3.6 | 10.7 | 100 |
| No. 4 | 0.8 | 10.4 | 100 |
| No. 5 | 2.2 | 1.4 | 10 |
| No. 6 | 2.2 | 1.2 | 4 |
| No. 7 | 2.8 | 1.4 | 4 |
| No. 8 | 1.2 | 1.1 | 4 |
| No. 9 | 0.44 | 0.06 | 0.7 |

Table 1. P1 center concentration and NV center concentration of HPHT samples.
