

Electrical Control for Extending the Ramsey Spin Coherence Time of Ion-Implanted Nitrogen-Vacancy Centers in Diamond

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The extension of spin coherence times is a crucial issue for quantum information and quantum sensing. In solid-state systems, suppressing noise through various techniques has been demonstrated. On the other hand, an electrical control for suppression is important toward individual controls of on-chip quantum-information devices. Here, we show electrical control for extension of the spin coherence times of 40-nm-deep ion-implanted single-nitrogen-vacancy center spins in diamond by suppressing magnetic noise. We apply 120 V dc across two contacts spaced by 10 μm . The spin coherence times, estimated from a free-induction decay and a Hahn-echo decay, are increased up to about 10 times (reaching 10 μs) and 1.4 times (reaching 150 μs), respectively. From the quantitative analysis, the dominant decoherence source, depending on the applied static electric field, is elucidated. Electrical control for extension can deliver a sensitivity enhancement to the dc sensing of temperature, pressure, and electric (but not magnetic) fields, opening up an alternative technique in solid-state quantum-information devices.

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I. INTRODUCTION

In quantum sensing [1–21] and quantum-information science [22–26], long coherence times are crucially important. The improvement in sensitivity is inversely proportional to the square root of the coherence times [1–9]. Additionally, their increase directly improves quantum-memory times [1–5,22–25] and quantum-gate fidelity [26,27] in quantum-information devices. Among solid-state systems, a negatively charged nitrogen-vacancy (N-V) center in diamond is significantly interesting because it has a long coherence time [1–5,22] and high sensitivity with a nanometer-scale resolution [6–15]. It has been eagerly investigated to realize applications such as high-precision magnetic [6,7,16] and electric field sensing [8–11,17], thermometry [12,13], sensing in or on living cell [13–15], quantum metrology [18,19], pressure sensing [20,21], and quantum-information processing [22–25].

Among the techniques to produce N-V centers for quantum sensing, the ion-implantation technique is the most effective way to deliberately put N-V centers very close to the surface with nanoscale resolution [28], which is needed

for nanoscale sensing of matter outside the diamond, as well as to have the best spatial resolution for sensing [7]. However, it is well known that the ion-implanted N-V centers have much shorter coherence times compared with the best ones due to defects created during ion implantation [7,29,30]. Therefore, a technique to improve the spin coherence time is important.

Extension of the spin coherence time has been demonstrated by suppressing noise through various techniques, such as tailored dynamic decoupling [31–35], measurements at low temperature [3], measurements at high magnetic field [36], and decoupling by fast charge-state changes with high-power laser irradiation [22], in addition to removal of noise sources by growth techniques [1,2,4,5]. On the other hand, the realization of suppression by an electric field is important [37–39] because the electric field can be locally controlled in individual on-chip devices, such as a scalable quantum device in a dense array [37–39] and a quantum sensing device. Furthermore, it does not need huge energy consumption facilities for operation and rare materials synthesized by isotopes without a nuclear spin.

Electrical control is challenging because the electric fields do not couple directly to the spin, unlike the

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magnetic field. Previously, the static electric field dependence on magnetic resonance frequencies of the N-*V* center was reported [17]. By using the dependence, application of the N-*V* center to nanoscale electric field sensors was demonstrated [8,10]. In previous research [8], analysis of the electronic structure of the N-*V* center reveals how an applied magnetic field influences the electric-field-sensing properties. Here, coherence times, which are estimated from a free-induction decay (T_2^{FID}) and a Hahn-echo decay (T_2^{echo}), are measured in the externally applied static electric field. We report the increase of T_2^{FID} and T_2^{echo} under the electric field and discuss the mechanism.

II. EXPERIMENTAL METHOD

The N-*V* center is a well-studied defect in diamond. It consists of a substitutional nitrogen atom adjacent to a vacancy site. The spin triplet of the ground state exhibits zero-field splitting of $D_{\text{g.s.}}/h \approx 2.87$ GHz between $m_s = 0$ and degenerate $m_s = \pm 1$ spin sublevels, where h is the Planck constant. The spin state of the triplet can be polarized and read out by an optical excitation and a photoluminescence (PL) intensity measurement. Owing to these properties, the electron-spin resonance of the N-*V* center can be sensed by optically detected magnetic resonance (ODMR) techniques.

In our experiment, N-*V* centers are created by ion implantation and subsequent annealing on an Ila (100)

single-crystalline diamond substrate. Nitrogen-14 (^{14}N), with a natural abundance concentration (99.6%), is implanted at a depth of about 40 nm by ion implantation with a kinetic energy of 30 keV. Electrodes and an antenna, to apply an electric field and microwaves, respectively, are formed on the substrate [Fig. 1(a)]. From the current-voltage property of the structure to apply an electric field [Fig. 1(b)], it is supposed that the voltage at the interface between the diamond and the electrode is zero and the electric field strength in diamond bulk is linearly proportional to the applied electric field (see Appendix A). The direction and strength of the electric field in diamond are simulated numerically [Fig. 1(c)]. The dots marked by “N-*V*1”, “N-*V*2”, and “N-*V*3” indicate the positions of the N-*V* centers. The estimated direction of the electric field for each N-*V* center is almost parallel to the y axis with a polar angle, θ_E , of 89° or 91° and an azimuth angle, φ_E , of 90° or 270° in the N-*V* coordinate system [Fig. 1(d)]. As shown in Fig. 1(e), the N-*V* axes of N-*V*1, N-*V*2, and N-*V*3 are parallel to one of the following axes: $[\bar{1}\bar{1}1]$, $[111]$, $[\bar{1}\bar{1}\bar{1}]$, or $[\bar{1}\bar{1}\bar{1}]$.

III. RESULTS AND DISCUSSION

Figure 2(a) shows the magnetic resonance spectra of N-*V*1 obtained by ODMR, while Fig. 2(b) plots the resonance frequencies from the fit with six Gaussian functions. Since six resonance signals are observed at each electric field, it

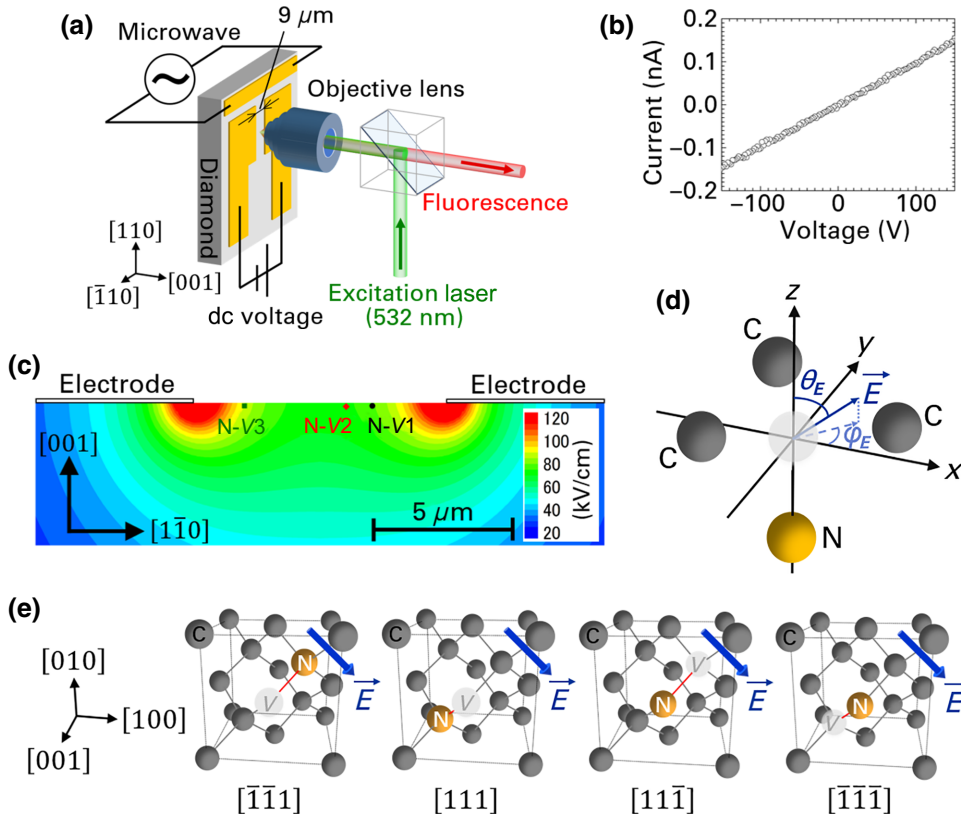


FIG. 1. (a) Schematic image of the measurement setup. (b) Current-voltage property between the electrodes on the sample. Linear I - V property is observed. (c) Distribution of the electric field strength with 100 V between the electrodes. Dots marked as N-*V*1, N-*V*2, and N-*V*3 indicate the positions of the N-*V* centers of N-*V*1, N-*V*2, and N-*V*3, respectively. (d) Coordinate system of the N-*V* centers. (e) Schematic images of the directions of N-*V* axes. N-*V* axes of N-*V*1, N-*V*2, and N-*V*3 are parallel to one of the following axes: $[\bar{1}\bar{1}1]$, $[111]$, $[\bar{1}\bar{1}\bar{1}]$, or $[\bar{1}\bar{1}\bar{1}]$.

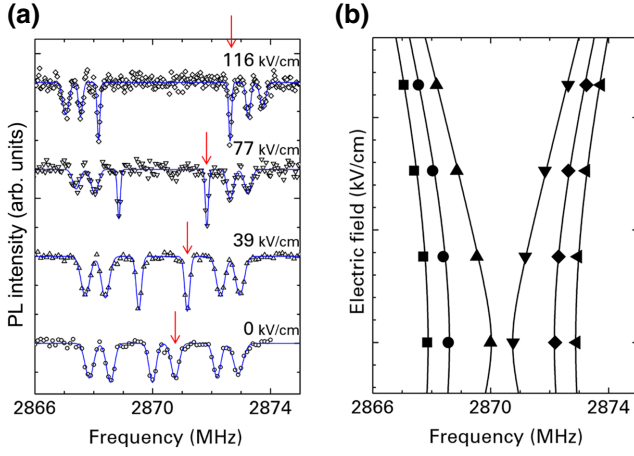


FIG. 2. (a) ODMR spectra under various electric fields. Plots represent the experimental results of N-V1. Solid lines represent the fitted curves using six Gaussian functions. (b) Electric field dependence of the resonance frequencies. Plots represent the estimated resonance frequencies from the experimental results of N-V1. Solid lines represent the fitted curves calculated from the spin Hamiltonian of the ground-state electron, Eq. (1), with the hyperfine interaction and quadrupole interaction of ^{14}N , Eq. (2).

seems that the N-V electron spin interacts with a nuclear spin of ^{14}N ($I = 1$). Thus, the z component of the external magnetic field, B_z , can be estimated from the obtained resonance frequencies at zero electric field. The estimated B_z values for N-V1, N-V2, and N-V3 are 13, 12, and 11 μT , respectively. The magnetic fields are likely from a residual field and/or the Earth's magnetism. As we increase the electric field, the spectral width becomes narrower and the resonance frequency shift becomes larger. From the narrowing of the spectral width, an increase in the coherence time by the electric field is inferred. Regarding the shifts in the resonance frequencies by the electric field, fittings based on the spin Hamiltonian of the ground triplet state are used in the quantitative analysis. The Hamiltonian is expressed as

$$H_{\text{g.s.}} = \frac{1}{\hbar^2} (D_{\text{g.s.}} + d_{\text{g.s.}}^{\parallel} E_z) \left[S_z^2 - \frac{1}{3} S(S+1) \right] + \frac{1}{\hbar^2} d_{\text{g.s.}}^{\perp} [E_x (S_y^2 - S_x^2) + E_y (S_x S_y + S_y S_x)] + \frac{1}{\hbar} \mu_B g_e \mathbf{S} \cdot \mathbf{B}, \quad (1)$$

where $\hbar$, μ_B , and g_e are the reduced Planck constant, the Bohr magneton, and the g factor of the electron spin, respectively. $\mathbf{S}$ is a spin operator and the spin quantum number of the N-V center is one ($S = 1$). $d_{\text{g.s.}}^{\parallel}/h = (0.35 \pm 0.02) \text{ kHz cm/kV}$ and $d_{\text{g.s.}}^{\perp}/h = (17 \pm 3) \text{ kHz cm/kV}$ [17] are the measured axial and non-axial components of the ground triplet-state electric dipole moment, respectively. To consider the effect of the ^{14}N

nuclear spin, the Hamiltonian describing the hyperfine interaction with the nuclear spin and the quadrupole interaction of the ^{14}N is introduced into the spin Hamiltonian, Eq. (1). The introduced Hamiltonian is given as

$$H_{\text{hfc}} = \frac{1}{\hbar^2} [A_{\parallel} S_z I_z + A_{\perp} (S_x I_x + S_y I_y) + P I_z^2], \quad (2)$$

where $A_{\parallel}/h = -2.1 \text{ MHz}$ and $A_{\perp}/h = -2.7 \text{ MHz}$ [40] are the axial component and the nonaxial component of the hyperfine interaction, respectively. I represents the nuclear spin of ^{14}N , and $P/h = -5.0 \text{ MHz}$ [39] is the strength of the quadrupole interaction of the ^{14}N nucleus.

The resonance frequencies at each electric field are determined by a numerical calculation and fitted to experimental results, where the free parameter of the fitting is the nonaxial components of the electric dipole moment $d_{\text{g.s.}}^{\perp}$. B_z is set to the estimated value from the spectrum at zero electric field. B_x and B_y are set to zero because their contributions are negligible under the conditions of $D_{\text{g.s.}} \gg \mu_B g_e B$ and $D_{\text{g.s.}} \gg d_{\text{g.s.}}^{\perp} E_{\perp}$.

Figure 2(b) shows the fitted curves as a solid line. The curves fit well with the experimental results, and $d_{\text{g.s.}}^{\perp}$ is estimated to be $d_{\text{g.s.}}^{\perp}/h = 19, 16, \text{ and } 16 \text{ kHz cm/kV}$ for N-V1, N-V2, and N-V3, respectively. The estimated values agree with the measured value of $d_{\text{g.s.}}^{\perp}/h = (17 \pm 3) \text{ kHz cm/kV}$ within the margin of error. These results indicate that the spin Hamiltonian given by Eq. (1) is valid in the range of a high electric field to about 100 kV/cm for the nonaxial component of the electric field effect on the ground-state electron spin of the N-V center. Additionally, we measure the shifts of the resonance frequencies at a range of about $\pm 100 \text{ kV/cm}$. The shifts behave as even functions with respect to the applied electric field. The result, indicating no electric-field-polarity dependence of the shifts, is consistent with the spin Hamiltonian (see Appendix B).

It is worth mentioning that, in Figs. 2(a) and 2(b), the hyperfine splitting observed in the ODMR seems to be weaker, as we increase the amplitude of the applied electric fields. This effect can be understood as follows. The hyperfine coupling effect on the N-V center from the nuclear spin can be interpreted as the effective Zeeman energy induced by magnetic fields from the nuclear spin. However, under the electric fields, the Zeeman splitting due to the magnetic fields will be suppressed. This means that the hyperfine splitting is also suppressed by applying the strong electric fields.

In previous research [41], it is shown that the frequency shifts of the magnetic resonance become dependent on the transverse orientation of the electric and magnetic field (i.e., ϕ_E and ϕ_B , where $\tan \phi_E = E_y/E_x$ and $\tan \phi_B = B_y/B_x$), if the electric and magnetic fields perpendicular to the N-V axis ($E_{\perp}$ and $B_{\perp}$) are simultaneously applied. They derive the equation for this dependence for the case that $E_{\perp}$

and $B_{\perp}$ are simultaneously applied, which is valid in the limit $\mu_B^2 B_{\perp}^2 / 2D_{\text{g.s.}} \gg d_{\text{g.s.}}^{\perp} E_{\perp}$. In our research, the geomagnetic field is unintentionally applied, although we do not know the direction of the geomagnetic field with respect to the N-V axis. Even if its direction is in the transverse plane, the applied electric field in our experiment is strong enough to satisfy the relation $\mu_B^2 B_{\perp}^2 / 2D_{\text{g.s.}} \ll d_{\text{g.s.}}^{\perp} E_{\perp}$. As shown in their paper, in the presence of either an electric field or a magnetic field, the resonance frequencies depend on the alignment of the electric and magnetic field with the center's major symmetry axis (i.e., θ_E and θ_B), but do not depend on the transverse orientation of the electric and magnetic field (i.e., ϕ_E and ϕ_B), where $\tan\theta_E = E_{\perp}/E_z$ and $\tan\theta_B = B_{\perp}/B_z$. Therefore, in our experiment, we can assume that the resonance frequencies do not depend on ϕ_E and ϕ_B .

To measure the coherence time, the microwave frequency is set to the arrowed transition shown in Fig. 2(a) and applying a weak microwave selectively excites the resonance. The resonance is related to the ^{14}N nuclear spin state of $m_I = 0$, where the effect of the hyperfine interaction on the resonance frequency is quite small. In this study, we characterize two kinds of coherence times: T_2^{FID} and T_2^{echo} . To measure the FID and the Hahn-echo signal, the Ramsey sequence and the Hahn-echo sequence, as depicted in Figs. 3(a) and 4(a), respectively, are employed. Figures 3(b) and 4(b) show the decay curves of the PL intensity as a function of τ , corresponding to the loss of coherence obtained from N-V1. Both T_2^{FID} and T_2^{echo} increase as the external electric field strength increases.

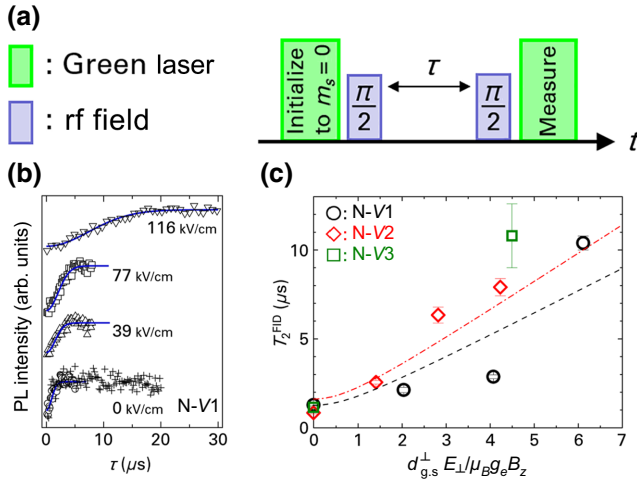


FIG. 3. (a) Ramsey pulse sequence to measure T_2^{FID} . (b) Decay curves of the free-induction decay of N-V1. Fit function of $f^{\text{FID}}(\tau) = y_0 + A \exp[-(\tau/T_2^{\text{FID}})^2]$ is employed. In 0 kV/cm, two data are represented as circles and pluses. (c) Electric field dependence of T_2^{FID} . Horizontal axis, which represents the electric field strength, is normalized by the magnetic field along the z axis. Dashed curve represents the fitted curve for N-V1. Chained curve represents the fitted curve for N-V2.

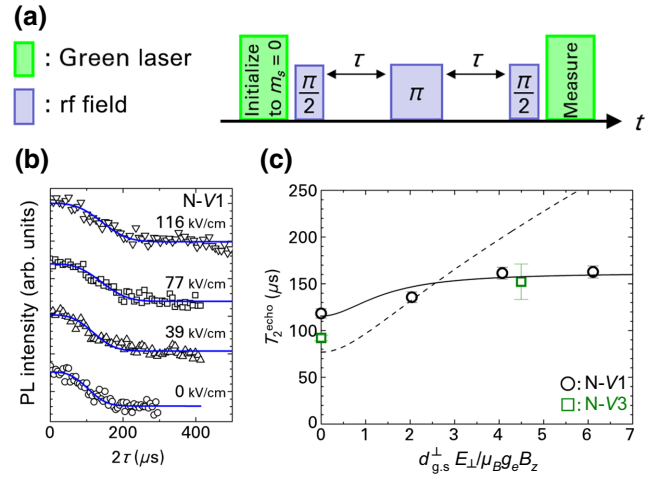


FIG. 4. (a) Hahn-echo pulse sequence to measure T_2^{echo} . (b) Decay curves of the Hahn-echo signal of N-V1. Fit function of $f^{\text{echo}}(2\tau) = y_0 + A \exp[-(2\tau/T_2^{\text{echo}})^3]$ is employed. (c) Electric field dependence of T_2^{echo} . Horizontal axis, which represents the electric field strength, is normalized by the magnetic field. Dashed curve represents the fitting curve with magnetic field fluctuation only. Solid curve represents the fitting curve with magnetic and electric field fluctuations.

T_2^{FID} and T_2^{echo} increase up to about 10 and 1.4 times, respectively. The decay curves of the free-induction decay of N-V2 and N-V3 are shown in Appendix C.

The dynamics of the N-V electron spin in bulk diamond are influenced by the locally fluctuating magnetic field induced by the surrounding spins (the spin bath) [2,42]. The impact of the spin bath on a N-V electron spin is limited to dephasing due to a fluctuating magnetic field along the N-V's quantization axis [31,36]. The magnetic field noise is represented by adding an effective noise term of $\mu_B g_e b_z(t)$, where $b_z(t)$ denotes the magnetic field from the spin bath. We assume that the spin bath can have temporal fluctuations, and thus, we consider $b_z(t)$ as random magnetic fields that stochastically change in time. We model the magnetic field fluctuation by the Ornstein-Uhlenbeck (O-U) process with the autocorrelation function, $C^b(t) = b_z(0)b_z(t) = b_z^{\sigma} 2 \exp(-|t|/\tau_c^b)$, where τ_c^b is the correlation time and $b_z^{\sigma} 2$ is the standard deviation of the amplitude distribution of the magnetic field fluctuation. The details of the spin bath are described in Appendix D. In the regime of $\sqrt{(\mu_B g_e B_z)^2 + (d_{\text{g.s.}}^{\perp} E_{\perp})^2} \gg \mu_B g_e b_z^{\sigma}$, which corresponds to our experiments that have a strong bias field or strong transverse electric field, a fluctuation in the resonance frequency $\delta\omega(t)$ induced by the magnetic field fluctuation under an electric field is expressed as

$$\delta\omega(t) = \sqrt{\{\mu_B g_e [B_z + b_z(t)]\}^2 + (d_{\text{g.s.}}^{\perp} E_{\perp})^2} - \sqrt{(\mu_B g_e B_z)^2 + (d_{\text{g.s.}}^{\perp} E_{\perp})^2} \cong R^b \frac{\mu_B g_e b_z(t)}{\hbar}, \quad (3)$$

where $R^b(E_\perp) = \mu_B g_e B_z / \sqrt{(\mu_B g_e B_z)^2 + (d_{g.s.}^\perp E_\perp)^2}$. T_2^{FID} and T_2^{echo} are estimated by the fit with functions of $f^{\text{FID}}(\tau) = y_0 + A \exp[-(\tau/T_2^{\text{FID}})^2]$ for the FID and $f^{\text{echo}}(2\tau) = y_0 + A \exp[-(2\tau/T_2^{\text{echo}})^3]$ for the Hahn echo, which are characteristics of a slowly fluctuating spin bath with $\hbar/(R^b \mu_B g_e b_z^\sigma) \ll \tau_c^b$ (see Appendix E). From these equations, we obtain the curves by fitting the data. Although there is a gap between data and curves because of the scattering of data; the obtained fitted curves are represented as blue solid lines in Figs. 3(b) and 4(b), while the estimated T_2^{FID} and T_2^{echo} are plotted in Figs. 3(c) and 4(c), respectively. Notably, in Figs. 3(b) and 4(b), the electric field strength is normalized by the magnetic field along the z axis (i.e., the horizontal axis represents $d_{g.s.}^\perp E_\perp / \mu_B g_e B_z$). Both T_2^{FID} and T_2^{echo} increase in accordance with the increase in the applied electric field.

To confirm the mechanism for the increase of the coherence time, we fit the functions

$$T_{2,b_z}^{\text{FID}}(E_\perp) = \frac{\sqrt{2}\hbar}{R^b \mu_B g_e b_z^\sigma}, \quad (4)$$

$$T_{2,b_z}^{\text{echo}}(E_\perp) = \left[12\tau_c^b \left(\frac{\hbar}{R^b \mu_B g_e b_z^\sigma} \right)^2 \right]^{\frac{1}{3}}, \quad (5)$$

to the experimental results of T_2^{FID} and T_2^{echo} with respect to the electric field, where the free parameter is b_z^σ for the fit of T_2^{FID} and τ_c^b for the fit of T_2^{echo} . b_z^σ in the fit of T_2^{echo} is set to the value estimated in the fit of T_2^{FID} . Notably, we suppose b_z^σ and τ_c^b are independent of the applied electric field. The fitted curves are represented as the dashed curves for N-V1 and the chained curve for N-V2 in Figs. 3(c) and 4(c), respectively. Regarding T_2^{FID} , the fitted curves basically reproduce the experimental results. The estimated b_z^σ values are 6 and 5 μT for N-V1 and N-V2, respectively.

In contrast to T_2^{FID} , the fitted curve for T_2^{echo} does not reproduce the experimental results. Particularly in the high-field region, the experimental result of T_2^{echo} is almost saturated. However, the fitted curve does not show saturation. Instead, the fitted curve increases, even at a high electric field. Therefore, another decoherence source should be considered in the analysis of T_2^{echo} .

For the additional decoherence source, the fluctuation in the electric field should be considered [43–46]. Although the influence of the fluctuation in the electric field on the resonance frequency becomes small in the regime of $\mu_B g_e B_z \gg d_{g.s.}^\perp E_\perp$ [45], the influence of the electric field fluctuation becomes larger than that of the magnetic field fluctuation in the regime of $\mu_B g_e B_z \ll d_{g.s.}^\perp E_\perp$. This behavior is consistent with the experimental results of T_2^{echo} .

Therefore, the fluctuation in the electric field is a candidate for the additional decoherence source. In the case of our system, the relevant fluctuations come from the y component of the electric fields due to the following reasons. First, the z component of the fluctuation can be ignored because the axial component of the electric dipole moment, $d_{g.s.}^\parallel$, is about 50 times smaller than that of the nonaxial component, $d_{g.s.}^\perp$. The x component can be ignored because the fluctuation in the nonaxial electric field, $E_\perp = \sqrt{E_x^2 + E_y^2}$, induced by the x component of the electric field fluctuation is suppressed in our experimental condition ($E_x \ll E_y$).

We model the electric field fluctuation, $e_y(t)$, by the O-U process with the autocorrelation function $C^e(t) = \langle e_y(0)e_y(t) \rangle = e_y^\sigma 2 \exp(-|t|/\tau_c^e)$, where τ_c^e is the correlation time and $e_y^\sigma 2$ is the standard deviation of the magnitude of the fluctuation. In addition, we assume that the fluctuations of the magnetic field and the electric field are not correlated. In the regime of $\sqrt{(\mu_B g_e B_z)^2 + (d_{g.s.}^\perp E_\perp)^2} \gg \mu_B g_e b_z^\sigma$, $d_{g.s.}^\perp e_y^\sigma$, the fluctuation in the resonance frequency $\delta\omega(t)$ due to the fluctuations of the magnetic and electric fields can be represented as

$$\begin{aligned} \delta\omega(t) = & \sqrt{\{\mu_B g_e [B_z + b_z(t)]\}^2 + \{d_{g.s.}^\perp [E_\perp + e_y(t)]\}^2} \\ & - \sqrt{(\mu_B g_e B_z)^2 + (d_{g.s.}^\perp E_\perp)^2} \cong R^b \frac{\mu_B g_e b_z(t)}{\hbar} \\ & + R^e \frac{d_{g.s.}^\perp e_y(t)}{\hbar}, \end{aligned} \quad (6)$$

where $R^e(E_\perp) = d_{g.s.}^\perp E_\perp / \sqrt{(\mu_B g_e B_z)^2 + (d_{g.s.}^\perp E_\perp)^2}$. Given the slow electric field fluctuation with $\hbar/[R^e d_{g.s.}^\perp e_y(t)] \ll \tau_c^e$, the electric field dependences of T_2^{FID} and T_2^{echo} are derived as

$$T_2^{\text{FID}}(E_\perp) = \frac{\sqrt{2}\hbar}{\sqrt{(R^b \mu_B g_e b_z^\sigma)^2 + (R^e d_{g.s.}^\perp e_y^\sigma)^2}}, \quad (7)$$

$$T_2^{\text{echo}}(E_\perp) = \frac{T_{2,b_z}^{\text{echo}} T_{2,e_y}^{\text{echo}}}{(T_{2,b_z}^{\text{echo}3} + T_{2,e_y}^{\text{echo}3})^{\frac{1}{3}}}, \quad (8)$$

where $T_{2,e_y}^{\text{echo}} = \{12\tau_c^e [\hbar/(R^e d_{g.s.}^\perp e_y^\sigma)]^2\}^{\frac{1}{3}}$. Regarding T_2^{FID} , it seems that the influence of fluctuation in the electric field is negligible compared with that of the magnetic field fluctuation, even at $E_\perp \sim 100 \text{ kV/cm}$, because the increase of T_2^{FID} by the electric field is not saturated. Thus, the fluctuation in the electric field cannot be estimated by fitting

with function (7), as the fitting is meaningless. However, we can roughly estimate the upper limit of the amplitude of the electric field fluctuation e_y^σ . We assume that the upper limit is the value satisfying the influence of the electric field fluctuation on T_2^{FID} , which is 10 times smaller than that of the influence of the magnetic field fluctuation [i.e., $(R^e d_{\text{g.s.}}^\perp e_y^\sigma)^2 \lesssim \frac{1}{10} (R^b \mu_B g_c b_z^\sigma)^2$]. The estimated value is $e_y^\sigma \lesssim 0.5$ kV/cm at $E_\perp \sim 100$ kV/cm.

Regarding T_2^{echo} , we fit function (8) to the experimental results with respect to the strength of the electric field, where the free parameters are τ_c^b and $\tau_c^e/e_y^\sigma 2$. Figure 4(c) shows the fitted curve as a solid line, reproducing the experimental results well. The estimated free parameters are $\tau_c^b = 170$ ms and $\tau_c^e/e_y^\sigma 2 = 6$ ms cm²/kV². From the estimated value and the upper limit of e_y^σ , the upper limit of τ_c^e is estimated to be 1.4 ms. The estimated values of the correlation times, $\tau_c^b = 170$ ms and $\tau_c^e < 1.4$ ms, are more than 10 times larger than that of the timescale of $T_2^{\text{echo}} \sim 0.1$ ms, which does not conflict with the assumptions of the slow fluctuations of the magnetic field and the electric field used in this analysis.

We discuss the source of electric field fluctuation. The reported surface-charge fluctuation [43,44,46] is a potential source of electric field fluctuation. Considering a point charge q on the surface, the resulting electric field at the N- V center in the diamond is expressed as $\mathbf{E} = (1/4\pi\epsilon_0)(2/[\kappa_d + \kappa_{\text{oil}}])(q/r^2)\hat{\mathbf{r}}$ [47], where the screening effect is not counted. r is the distance between the surface electron and the N- V center, and $\hat{\mathbf{r}}$ is the unit vector in the direction of the N- V center. $\kappa_d = 5.7$ and $\kappa_{\text{oil}} = 2.3$ are the dielectric constants of diamond and optical immersion oil, respectively. ϵ_0 is the permittivity of free space. For a single elementary charge located immediately above (40 nm) the N- V center, the strength of the electric field is about 2 kV/cm. Although the value is larger than the upper limit of e_y^σ estimated above, this result is not necessarily incongruous because the electric field is partially screened by the interplay of the electrons in the diamond lattice, maintaining electroneutrality. We consider that the origin of the electric field fluctuation may be the surface-charge fluctuation [44] or charge fluctuation of impurities and defects in bulk. Our results indicate that T_2^{echo} will increase almost linearly, as shown by the dotted line in Fig. 4 (c), by reducing the fluctuation.

In our case, the maximum applied electric field is limited by a breakdown of the electrode deposited on diamond. This upper limit will be improved technically by finding optimized fabrication processes and will be extended to the intrinsic breakdown voltage of diamond. One of the largest breakdown voltages of diamond is reported to be more than 2 MV/cm [48]. This is more than 10 times larger than the maximum applied electric field (166 kV/cm) in the present experiment. Therefore, in principle, a further extension of T_2^* can be expected.

IV. CONCLUSION

The electric field dependence of T_2^{FID} and T_2^{echo} over a range up to about 100 kV/cm are quantitatively analyzed based on the spin Hamiltonian. T_2^{FID} and T_2^{echo} increase up to about 10 and 1.4 times, respectively. Assuming only a magnetic field fluctuation, the behavior of T_2^{FID} is basically elucidated. The behavior of T_2^{echo} is well reproduced, assuming fluctuations in both the electric and magnetic fields. Although the magnetic field fluctuation is the dominant decoherence source for T_2^{FID} over the entire range of electric field in our experiment and for T_2^{echo} in the low-electric-field region, the dominant decoherence source for T_2^{echo} in the high-electric-field region is the electric field fluctuation. The difference in the dominant decoherence source under the electric field is due to the difference in the amplitude-frequency characteristics, corresponding to the difference in the correlation time of the magnetic field fluctuation and the electric field fluctuation in our experiment. The enhancement of the coherence times by the electric field can contribute to the improvement of the sensitivity in thermometry, pressure, and ac electric field sensing, although it is not effective on the magnetic field sensitivity [44]. The present technique can be utilized for not only for the N- V center in diamond, but also the high-spin centers ($S > 1$) with zero-field splitting, such as promising centers in silicon carbide [49–51]. Our study opens up an alternative technique for the electrical decoupling of spin coherence in a solid from magnetic noise.

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APPENDIX A: MATERIALS AND APPLICATION OF THE ELECTRIC FIELD

The diamond substrate is a CVD-grown Ila (100) single-crystalline diamond purchased from Element Six Corporation (electronic grade). The nitrogen impurity concentration is less than 5 ppb and the concentration of ¹³C is the natural abundance in diamond. ¹⁴N with a natural abundance concentration is ion implanted at the Ion Technology Center Co., Ltd., with a kinetic energy of 30 keV, implanting nitrogen at a depth of about 40 nm with a density of about 5×10^8 /cm². Subsequent annealing at 800 °C for 1 h, using rapid thermal annealing equipment, produces N- V centers with a density around less than 0.1 ppb. To remove residual surface contamination and terminate the surface with oxygen, the sample is kept in a mixture of H₂SO₄

and HNO_3 at 200 °C for 1 h. Ti(30 nm)/Au(100 nm) electrodes for the electric field and an antenna for microwaves are formed on the substrate by conventional electron-beam lithography and metal-deposition processes.

The linear current-voltage (I - V) property of the structure for applying an electric field is obtained, although there are Schottky barriers at the interface between the electrodes and diamond. Schottky barriers exhibit a linear I - V property in the regime of $V \ll |kT/e|$, where k is the Boltzmann constant, T is the temperature, and e is the elementary charge. Thus, in the range of the applied electric field, the voltage at the interfaces appears to be much smaller than that of $|kT/e|$ (approximately 0.026 V at room temperature). The voltage at the interface is negligible compared with the voltage applied to the electrodes. Therefore, we assume that the voltage at the interface is zero and the electric field strength in diamond bulk is linearly proportional to the applied electric field.

The direction and strength of the electric field in diamond is simulated numerically [Fig. 1(c)]. The dots marked by “N- $V1$ ”, “N- $V2$ ”, and “N- $V3$ ” indicate the positions of the N- V centers of N- $V1$, N- $V2$, and N- $V3$, respectively. Their positions are estimated from the confocal-scan images and the depth of implanted nitrogen. The direction of the N- V axes of the N- V centers are parallel to one with $[111]$, $[\bar{1}\bar{1}\bar{1}]$, $[1\bar{1}\bar{1}]$, or $[\bar{1}11]$, which is confirmed by the measurements of PL intensity with polarized light. From the results, the direction of the electric field in the N- V coordinate system depicted in Fig. 1(d) is estimated to be almost parallel to the y axis with a polar angle, θ_E , of 89° or 91° and an azimuth angle, φ_E , of 90° or 270°.

APPENDIX B: ELECTRIC-FIELD-POLARITY DEPENDENCE OF THE RESONANCE FREQUENCY SHIFTS AND THE TRANSITION RATES

We measure the electric-field-polarity dependence of the resonance frequency shifts at a range of about ± 100 kV/cm because asymmetry of the optical transition, depending on the polarity, was reported previously [52]. The measured N- V center is another one, N- $V4$. The direction of the N- V axis of N- $V4$ is parallel to one with $[1\bar{1}\bar{1}]$, or $[\bar{1}11]$, which is different from that of N- $V1$ -3. Thus, the x component of the electric field is much larger than the y component. Figure 5(a) shows the magnetic resonance spectra of N- $V4$ obtained by ODMR. While four resonance signals are observed at zero electric field, the two dips in the middle of the four dips are larger than those of the others. It seems that the two resonance signals overlap at the middle two dips and the N- V electron spin interacts with a nuclear spin of ^{14}N ($I = 1$). Then the z component of the external magnetic field, B_z , can be estimated from the obtained resonance frequencies at zero electric field. The estimated B_z

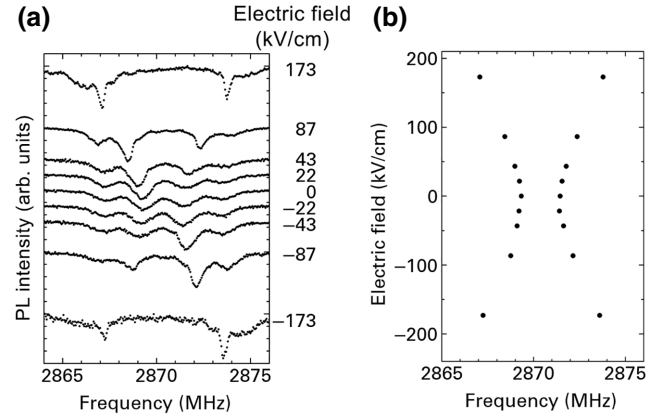


FIG. 5. (a) ODMR spectra under various electric fields. Plots represent the experimental results of N- $V4$. (b) Electric field dependence of the resonance frequencies. Plots represent the estimated resonance frequencies from the experimental results of N- $V4$.

value is 34 μT . Figure 5(b) plots the resonance frequencies of the middle two with respect to the electric field strength. The shifts behave as even functions with respect to the applied electric field. The result, indicating no electric-field-polarity dependence of the shifts, is consistent with the spin Hamiltonian.

Regarding the size of the dips, an electric-field-polarity dependence is observed. This may be due to a change of the magnetic transition rates. The transition rates are approximately described by Fermi's golden rule as $\frac{2\pi}{\hbar} |\langle S_{\pm} | H_{\text{MW}} | S_0 \rangle|^2$, where $|S_0\rangle$, $|S_+\rangle$, and $|S_-\rangle$ are eigenstates of the electron spin, which are expressed as $|S_0\rangle = |0\rangle$, $|S_+\rangle = e^{i(\varphi_E/2)} \cos \frac{\theta}{2} | +1 \rangle - e^{-i(\varphi_E/2)} \sin \frac{\theta}{2} | -1 \rangle$, and $|S_-\rangle = e^{i(\varphi_E/2)} \sin \frac{\theta}{2} | +1 \rangle + e^{-i(\varphi_E/2)} \cos \frac{\theta}{2} | -1 \rangle$, respectively. Here, $\tan \theta = (d_{\text{gs}}^{\perp} E_{\perp}) / (\mu_B g_e B_z)$ and $|0\rangle$, $|+1\rangle$, and $|-1\rangle$ are eigenstates of S_z . H_{MW} is the perturbing Hamiltonian due to the microwave. In our measurement system, the x component of the oscillating field from the microwave $B_{\text{MW},x}$ is much larger than that of the y component. Thus, the perturbing Hamiltonian is expressed as $H_{\text{MW}} = (1/\hbar) \mu_B g_e S_x B_{\text{MW},x}$. In the case where the electric field is along the y axis, the transition rates do not depend on the strength of the electric field. Actually, the observed Rabi frequencies of the N- $V1$ at each electric field are almost the same. On the other hand, in the case where the electric field is along the y axis, the transition rates depend on the strength of the electric field. This result agrees with the experimental results of N- $V4$ qualitatively.

APPENDIX C: DECAY CURVES OF THE FREE-INDUCTION DECAY OF N- $V2$ AND N- $V3$

The decay curves of the free-induction decay of N- $V2$ and N- $V3$ are shown in Figs. 6(a) and 6(b), respectively. The Ramsey pulse sequence to measure T_2^{FID} is the same

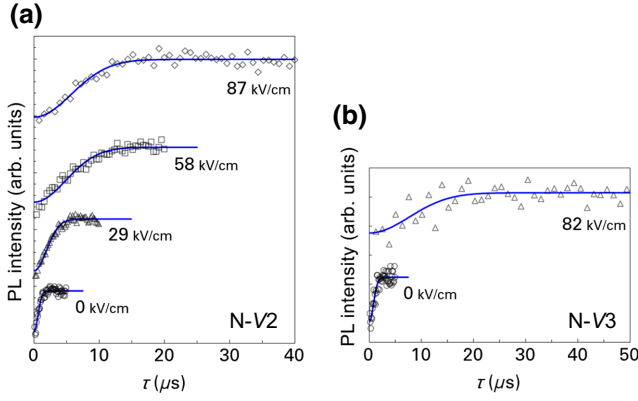


FIG. 6. (a) Decay curves of the free-induction decay of N-V2. Fit function of $f^{\text{FID}}(\tau) = y_0 + A \exp[-(\tau/T_2^{\text{FID}})^2]$ is employed. (b) Decay curves of the free-induction decay of N-V3. Fit function of $f^{\text{FID}}(\tau) = y_0 + A \exp[-(\tau/T_2^{\text{FID}})^2]$ is employed.

as that of N-V1, as shown in Fig. 3(a). The fit function of $f^{\text{FID}}(\tau) = y_0 + A \exp[-(\tau/T_2^{\text{FID}})^2]$ is employed. The estimated T_2^{FID} values are plotted in Fig. 3(c).

APPENDIX D: CONSIDERATION OF THE SPIN BATH

The main decoherence source of the N-V centers located in bulk diamond is the fluctuation in the magnetic field induced by paramagnetic centers in the lattice [2,42]. For the N-V centers situated near the surface (<10 nm), the main sources of decoherence are the fluctuations in the magnetic field due to the flip of electron spins at the surface [29] and the fluctuations in the electric field due to a charge fluctuation in the surface states [43,44]. In our experiments, the N-V centers are placed around 40 nm from the surface. Therefore, the fluctuation in the magnetic field from paramagnetic centers should be the main source of decoherence. With regard to the paramagnetic centers, the nuclear spin of ^{13}C atoms and the electron spins of some types of defects (e.g., N-substituted defects) are possible candidates. For a case in which naturally abundant ^{13}C (1.1%) is a main source of decoherence, T_2^{echo} for the N-V electron is about 500 μs [2].

In our experiment, the timescale of the decay curves of the Hahn-echo measurement is about 100 μs . The short timescale in our experiments indicates that the additional contribution to decoherence is attributed to the electron spins, constituting a spin bath. Since the magnetic moments of the electron spins in the spin bath and N-V centers are similar in size, there is no significant difference in the coupling strength of between intrabath spin-spin interaction and bath spin to the N-V electron-spin interaction. In such a situation where many spins interact with each other with similar coupling strengths, the effect of the N-V electron spin to the spin bath diffuses to the bath rapidly. This behavior is similar to the individual spins

in the bath, allowing fluctuation in the magnetic field to be treated as stochastic noise made by a classical Markov process [31,36]. Additionally, under the assumption of a uniform distribution of defects in the spin bath, fluctuation in the magnetic field is expressed by a Gauss distribution. In summary, the magnetic field fluctuation, according to the stationary O-U process, is assumed in this study.

Although the fluctuation in the magnetic field is three dimensional, only the z component affects fluctuation in the resonant frequency and decoherence. The x and y components flip the N-V electron spin, in principle. However, a large difference in the spin-flipping energy between the N-V center and other defects due to a zero-field splitting of 2.87 GHz in the N-V center results in negligible spin-flip probabilities [31,36]. Here, we denote the z component of the fluctuating magnetic field as $b_z(t)$. From the aforementioned assumptions, the distribution of $b_z(t)$ is Gaussian with an average value of $b_z(t) = 0$. The autocorrelation function, $C^b(t)$, is $C^b(t) = b_z(0)b_z(t) = b_z^2 \exp(-|t|/\tau_c^b)$. Here, τ_c^b is the correlation time and b_z^2 is the standard deviation in a fluctuating magnetic field. It should be noted that the correlation time, τ_c^b , and standard deviation, b_z^2 , correspond to the strength of the spin-spin interaction inside the spin bath and the N-V center to the bath-spin interaction, respectively.

APPENDIX E: THE ANALYSIS OF T_2^{FID} AND T_2^{echo}

To derive the fitted functions for the electric field dependence of T_2^{FID} and T_2^{echo} , the time evolutions of the spin in the Ramsey and Hahn-echo sequences with the fluctuation in the resonance frequency should be calculated. The statistical average with respect to the fluctuations should be taken into account. The population of the N-V electron spin with $m_s = 0$ after free evolution is expressed as $p_{S_0}^{\text{FID}}(\tau) = (1/2) - (1/2)\text{Re}\{\exp[-i \int_0^\tau \delta\omega(t)dt]\}$ for the FID and $p_{S_0}^{\text{echo}}(2\tau) = (1/2) + (1/2)\text{Re}\{\exp[-i \int_0^\tau \delta\omega(t)dt]\exp[i \int_\tau^{2\tau} \delta\omega(t)dt]\}$ for the Hahn echo. In the case where the fluctuation in the resonance frequency is expressed by Eq. (3), $p_{S_0}^{\text{FID}}(\tau)$ and $p_{S_0}^{\text{echo}}(2\tau)$ are derived as $p_{S_0}^{\text{FID}}(\tau) = (1/2) - (1/2)\exp\{-\tau_c^b [R^b(\mu_B g_e b_z^\sigma/\hbar)]^2 [(\tau/\tau_c^b) - 1 + e^{-(\tau/\tau_c^b)}]\}$ and $p_{S_0}^{\text{echo}}(2\tau) = (1/2) + (1/2)\exp\{-\tau_c^b [R^b(\mu_B g_e b_z^\sigma/\hbar)]^2 [(2\tau/\tau_c^b) - 3 - e^{-(2\tau/\tau_c^b)} + 4e^{-(\tau/\tau_c^b)}]\}$, respectively. Given the slow magnetic field fluctuation with $\hbar/R^b \mu_B g_e b_z^\sigma \ll \tau_c^b$, $p_{S_0}^{\text{FID}}(\tau)$ and $p_{S_0}^{\text{echo}}(2\tau)$ are simplified to be $p_{S_0}^{\text{FID}}(\tau) = (1/2) - (1/2)\exp\{-(1/2)[R^b(\mu_B g_e b_z^\sigma/\hbar)]^2 \tau^2\}$ and $p_{S_0}^{\text{echo}}(2\tau) = (1/2) + (1/2)\exp\{-(1/12\tau_c^b)[R^b(\mu_B g_e b_z^\sigma/\hbar)]^2 (2\tau)^3\}$. From these equations, the functions describing the electric field dependence of T_2^{FID} and T_2^{echo} are derived as Eqs. (4) and (5). Equations (7) and (8) are derived by the same calculations.

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